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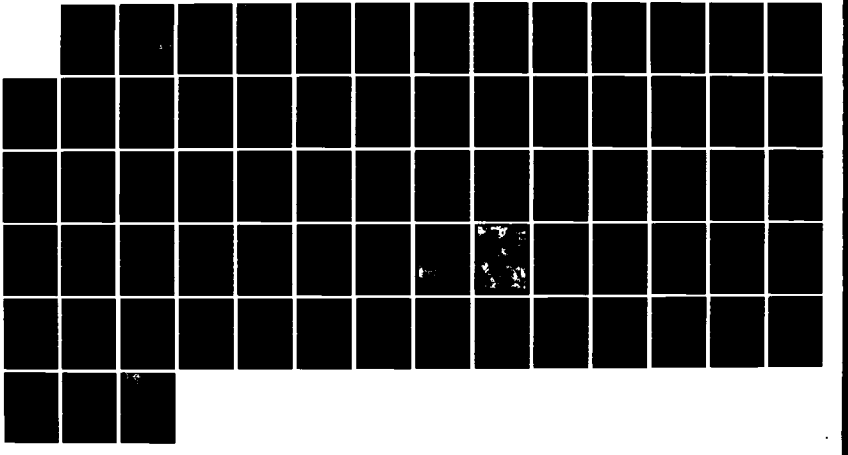

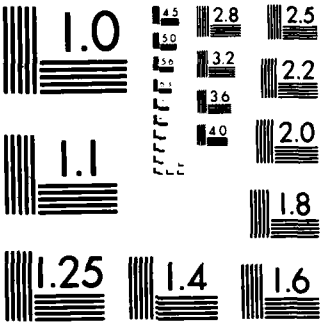

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Report No. 05423F

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Semiconductors Investigated by Time Resolved Raman Absorption and Photoluminescence Spectroscopy Using Femtosecond and Picosecond Laser Techniques

by

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

This report summarizes the research progress achieved in the period 1979-1982 in the research effort supported by AFOSR 80-0079. The main areas of research are: picosecond and subpicosecond laser development and application and time-resolved studies of semiconductors.

In the subpicosecond laser development program, we investigated a variety of cavities of different physical parameters. A

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19. generated Hot-carriers, Carrier Recombination, Semiconductors GaAs, GaSe (layered semiconductor) $CdCr_2Se_4$ (magnetic semiconductors).

20. stable and reliable oscillator, which produces 200 fsec pulses, has been developed using a ring configuration. The first generation amplifier system has been completed with a gain of 10^6. We have produced continuum by focusing the amplified pulses in a CCl_4 cell; and have invented a femtosecond luminescence technique (called population mixing) for probing semiconductor processes. In addition, we have improved the stability and shortened the pulse duration emitted from a mode-locked glass laser by at least a factor of two, by using heptamethine pyrylium #5 - a new saturable absorber.

In the semiconductor research program we have conducted a systematic investigation on $CdCr_2Se_4$, GaAs and GaSe using absorption and luminescence kinetic spectroscopy. In the magnetic semiconductor $CdCr_2Se_4$ our measurements have resolved a long standing controversy over the assignments of the valence band to conduction band transitions, the electronic band structure, and the size of the fundamental gap.

We have observed that the hot photogenerated carrier distribution in both GaAs and GaSe cools at a slower rate at high electron-hole plasma densities. The slow kinetics arise from the screening of the electron-optical-phonon (GaAs) and hole-optical-phonon (GaSe) interactions. The deformation potential was calculated for carrier-phonon interaction in GaSe. The photoexcited carrier recombination in p-type GaAs was measured using the population mixing technique and the subpicosecond laser. The recombination of $CdCr_2Se_4$ was measured to be less than 2 psec. The ultrafast recombination time may allow for the development of fastest optical switches at room temperature.

Over the grant period, 27 papers have been published in scientific journals, and 20 technical presentations at professional meetings have been given. The USL has trained 6 scientists in the art of ultrafast technology with AFOSR support.

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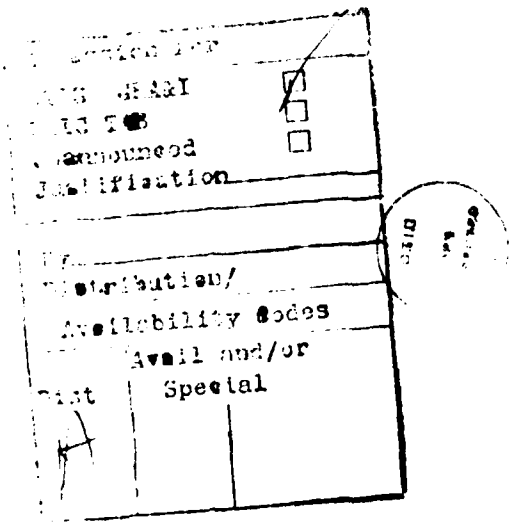

I. SUMMARY

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II. LIST OF PUBLICATIONS

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Population Mixing Correlation Subpicosecond/Picosecond Time Resolved Luminescence Technique for Semiconductors and other Multi-recombination Processes, R. R. Alfano, D. Rosen, A. Katz, Y. Budansky, A. Doukas (1982).

IV. FIRST YEAR PROGRESS REPORT

Considerable progress has been made during the first year of the grant with respect to: (1) the measurements of the relaxational kinetics of photo-excited carriers in the magnetic semiconductor $CdCr_2Se_4$ and in the semiconductor GaAs, and (2) the construction and characterization of the dye femtosecond laser. The next sections will summarize this research.

1) SEMICONDUCTOR RESEARCH

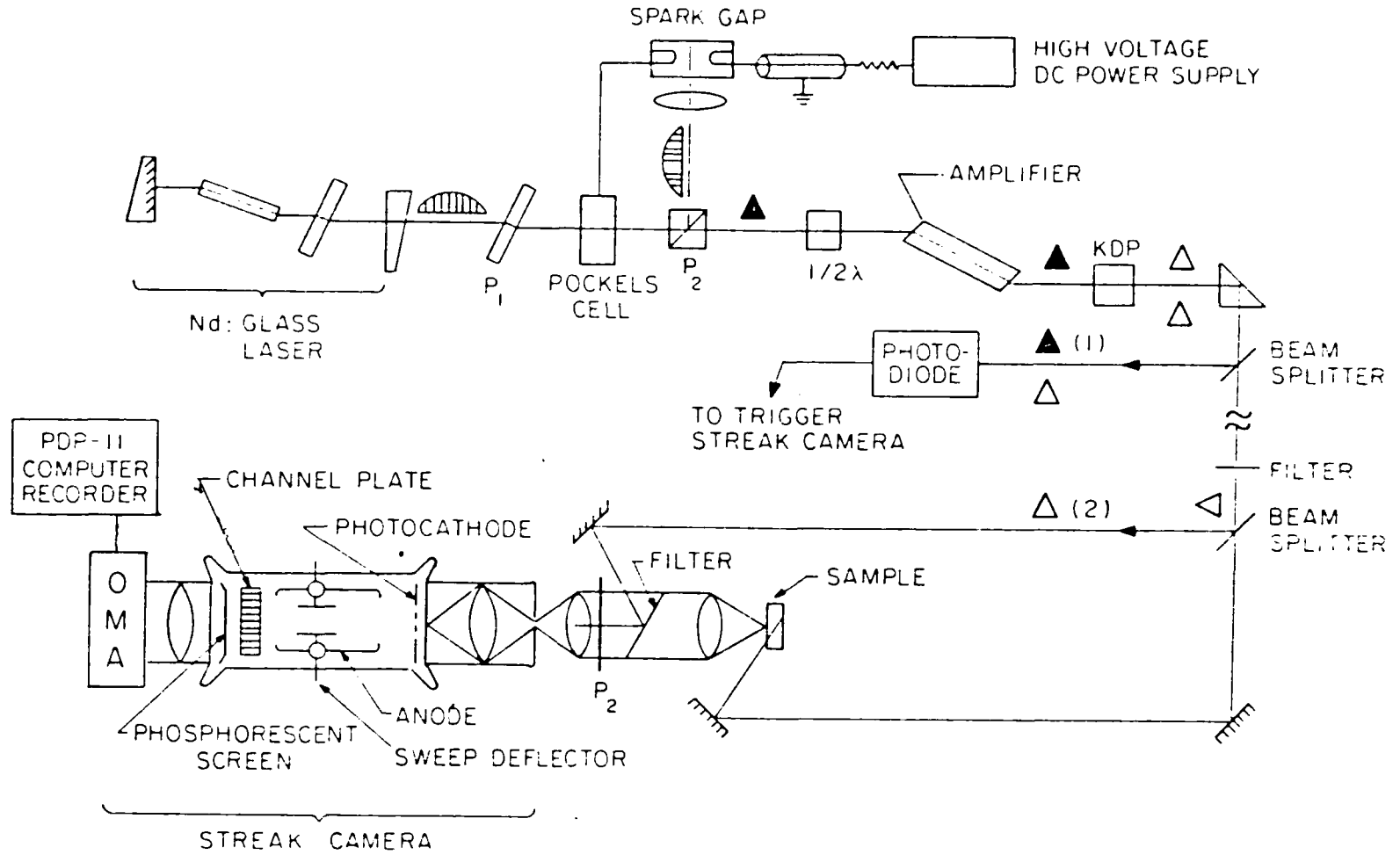
In this section, we will summarize our research on the photoluminescence kinetics $CdCr_2Se_4$ and GaAs.

The experimental setups used to measure the photoluminescence kinetics and CW spectra are shown in setups 1 and 2, respectively.

SETUP 1: In this experiment, a Nd:glass mode-locked laser was used to generate picosecond light pulses. A single pulse was selected from train using a Pockel cell. The pulse was amplified and converted in a potassium dihydrogen phosphate crystal to a second harmonic plse at 530 nm. The pulse duration at 530 nm is about 6 ps. The 530 nm pulse was collimated onto the crystal. A $1mm^3$ $CdCr_2Se_4$ crystal was held inside an optical dewar on an aluminum plate with a microscopic glass plate. The photon flux density in a pulse at the sample was about 10^{14} photons/cm^2.

The photoluminescent kinetics were measured using a streak camera in which photoelectrons produced at various times by the photons arriving at the photocathode are caused to streak across a phosphorescent screen to produce a temporal profile of the light emitting event. The streak camera used in this experiment was

manufactured by Hamamatsu. The prompt response time of the streak camera is about 10 psec. The photoluminescence was collected by an f/1.25 lens into the 100 μ slit of the streak camera. The intensity of the streak was recorded by a video optical multichannel analyzer (OMA by Princeton Applied Research) and an X-Y recorder. The intensity kinetic photoluminescent profiles were stored in a DEC PDP 11/03 computer for data analysis.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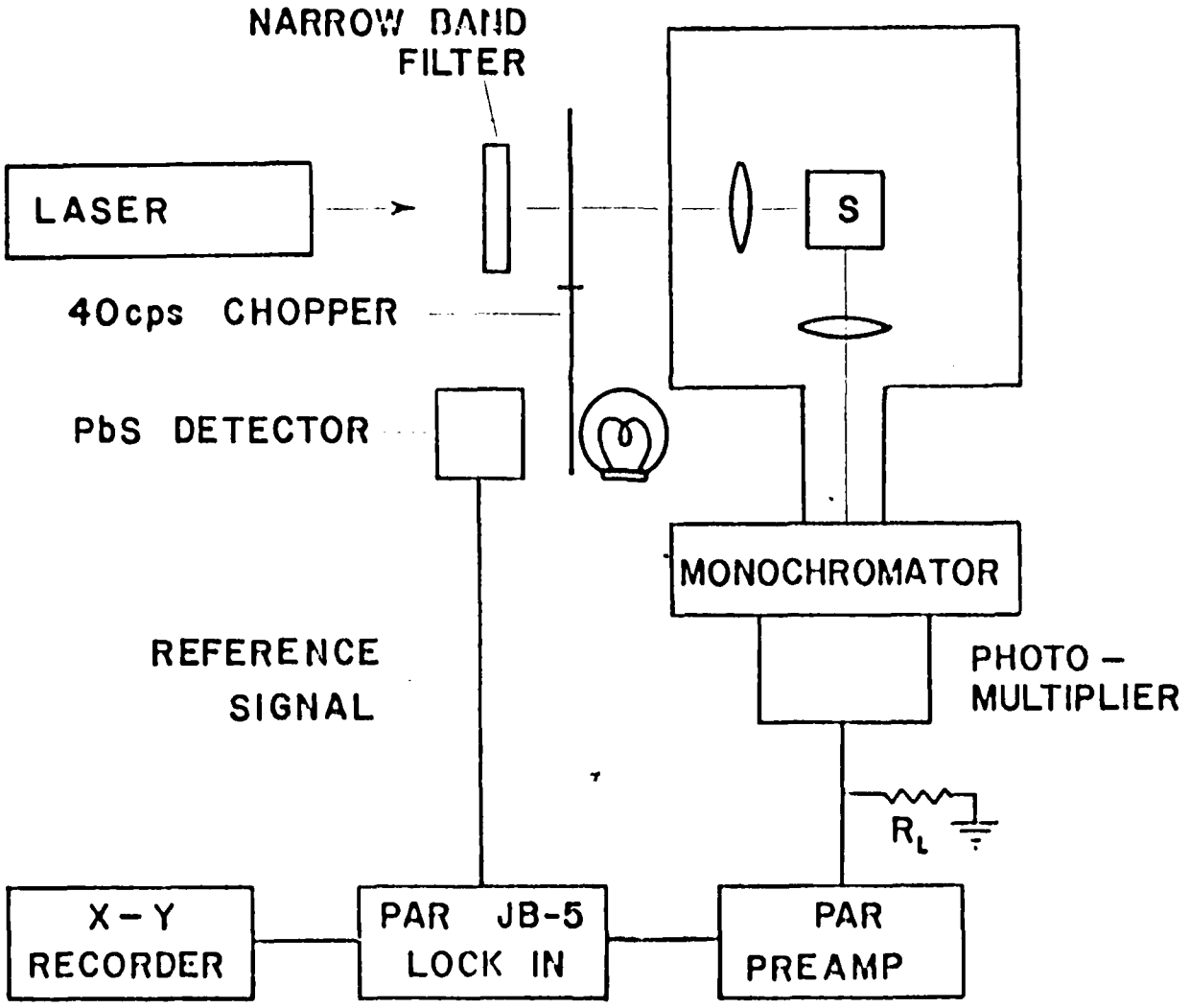
SETUP 2: The photoluminescent spectra was excited using a CW 5 mw Argon laser beam at 4880 Å. The argon laser beam was chopped at 100 Hz. The photoluminescent spectra was detected by a double]-m Spex spectrometer, a RCA 7265 photomultiplier tube, a lock-in amplifier, and a recorder.

LUMINESCENCE SETUP



Setup 2

A)- Magnetic Semiconductor $CdCr_2Se_4$

The ferromagnetic semiconductors have aroused considerable interest in novel applications which utilize both electrical and magnetic properties[6].

MAGNETICALLY SENSITIVE DIODE

MAGNETICALLY SENSITIVE TRANSISTOR

GUNN EFFECT MICROWAVE OSCILLATORS

GIANT MAGNET ORESISTANCE

ULTRAFAST MAGNETO-OPTIC COMPUTERS

ULTRAFAST OPTICAL SWITCHES

$CdCr_2Se_4$ orders ferrinagnetic at a high temperature and has attached special attention because it displays optically induced permeability changes[7] as well as magnetization changes.

The physical properties of $CdCr_2Se_4$ are as follows:

a. Magnetic Ordering: Ferromagnetism

   Curie Temperature: 130^oK

b. Saturation Magnetism: 5.7 μ_B (4.2^oK)

c. Optical Absorption Edge: 1.32 ev (Room Temp)

d. Crystal Structure: Spinel

e. Cell Dimension: 10.755 A

f. Semiconductors:

   n-type: In-doped

   p-type: Ag-doped

   conductivity: $3x10^3(\Omega cm)^{-1}$, p-type, 298^oK

   Hall mobility: 25 cm^2/v_{sec}, p-type, 298^oK

Pure $CdCr_2Se_4$ is always p-type with average carrier density of $10^{16}cm^{-3}$ at room temperature.

Recently, the band structure of $CdCr_2Se_4$ has been calculated .

The following is a list of measurements performed which summarizes our progress from 12/1/79 - to date:

1. The photoluminescent relaxation decay times of $CdCr_2Se_4$ have been measured as a function of the temperature using the picosecond laser and streak camera.
2. The photoluminescent spectra of temperature using a low power CW Argon laser.. The carrier temperatures have been calculated from the high energy tails of the spectra and compared with the sample temperature.
3. The peak energy and the linewidth of the photoluminescence from $CdCr_2Se_4$ have been measured as functions of the temperature using a low power CW Argron laser excitation.
4. The luminescent intensity of $CdCr_2Se_4$ as a function of the pumping intensity have been measured using a high power picosecond laser pulse at RT and a low power CW Argon at 77K.
5. The photoluminescent intensity of $CdCr_2Se_4$ as a function of temperature has been measured using a low power CW laser from 90K to 180K.

The following seven figures summarizes these measurements:

Fig. 1: The photoluminescent kinetics of $CdCr_2Se_4$ at 77K. The laser pulse at left is a calibrating pulse arriving at the streak camera at an earlier time. The time difference between the markers is 20 ps. The decay time of the luminescence is 14.7 ps.

Fig. 2: The recombination time of the photoluminescence of $CdCr_2Se_4$ versus temperature. The salient feature of this figure is that the decay time decreases when the temperature increases from 77K.

Fig. 3: The peak energy and linewidth of the photoluminescence of $CdCr_2Se_4$ versus temperature measured in ev using a low power CW Argon laser. The peak energy decreases and the linewidth increases when the temperature increases. The peak of the luminescence at 77K occurs at 6860 A. The luminescence blue shifts by 120 A when the temperature is lowered from 173K to 77K.

Fig. 4: The emission spectra of $CdCr_2Se_4$ at two different temperatures.

Fig. 5: The carrier temperatures calculated from the high energy tails of the luminescent spectra versus the sample temperatures.

Fig. 6: The luminescent intensity versus the low intensity of CW Argon at 77K. Theoretically, for intrinsic semiconductor the m value should be equal to 2 because the luminescent intensity is proportional to the excited electron and the hole concentration.

Fig. 7: The integrated luminescent intensity versus the pumping photon flux of the picosecond laser at room temperature.

Fig. 8: The luminescent intensity versus the temperature using a low power Argon CW laser of 5 mw. It is clear that below 130K - the Curie temperature of $CdCr_2Se_4$, the intensity starts to increase dramatically as the temperature is lowered.

Figure 1

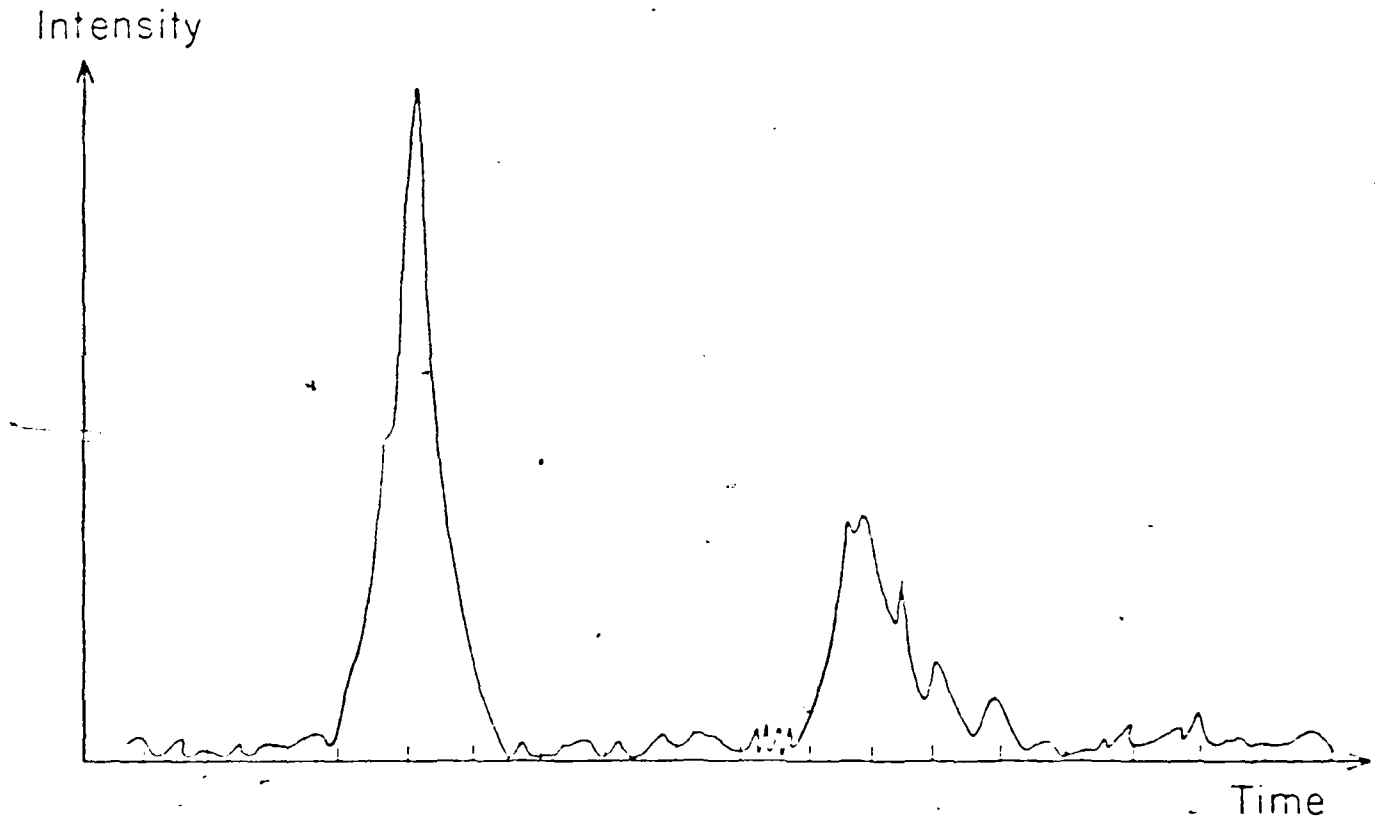
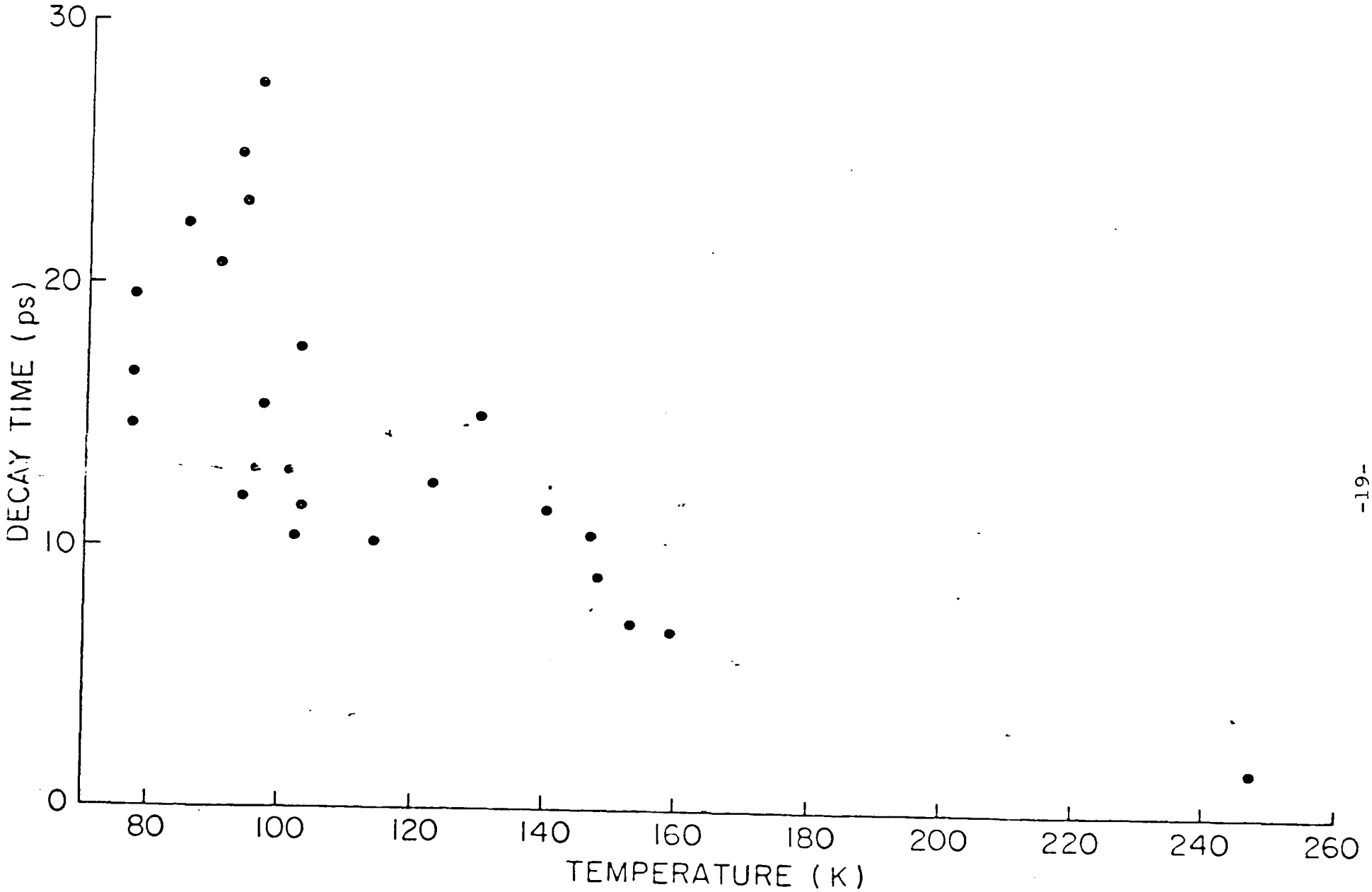


Figure 2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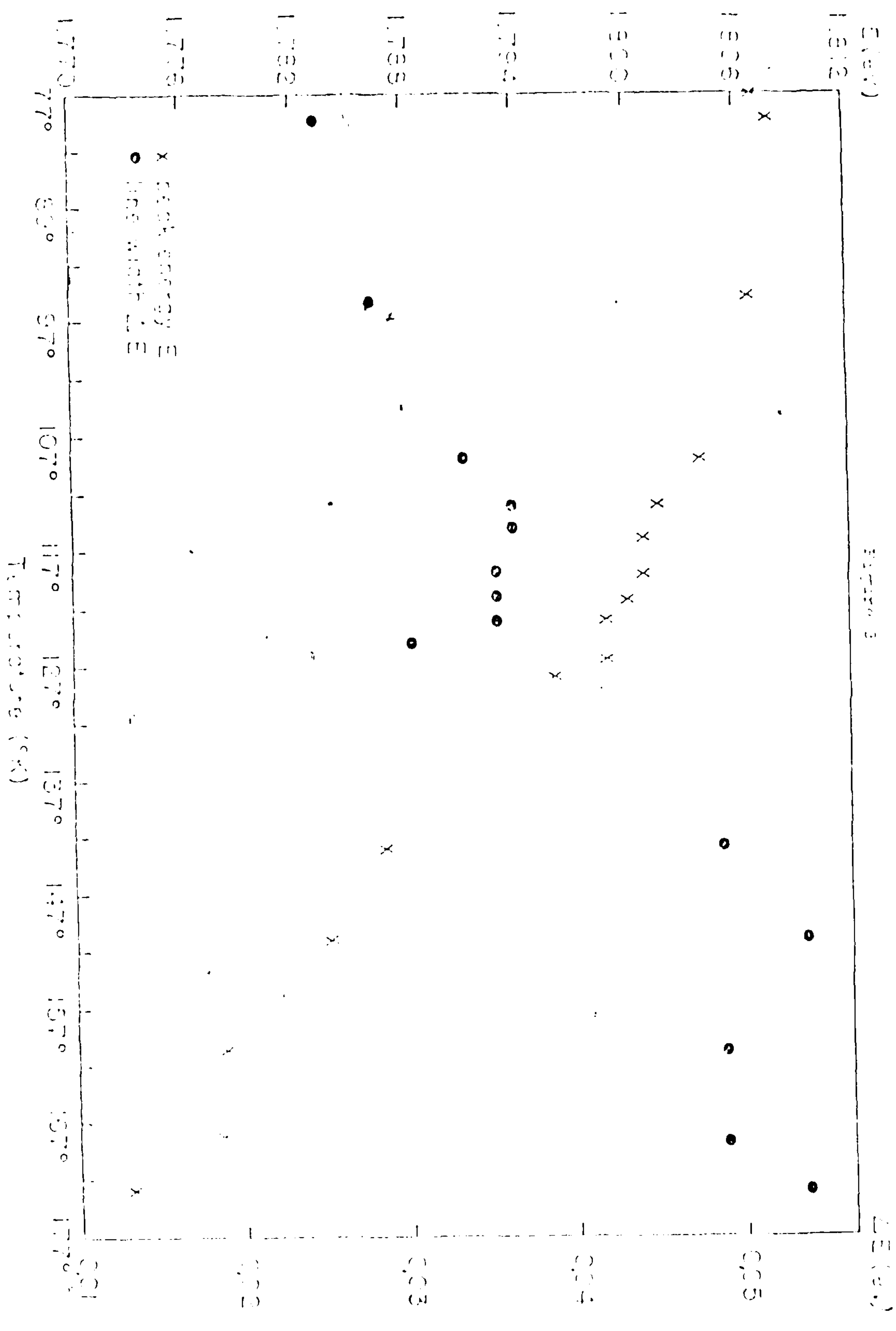


Emission Spectrum of $CdCr_2Se_4$ at Two Different Temperatures

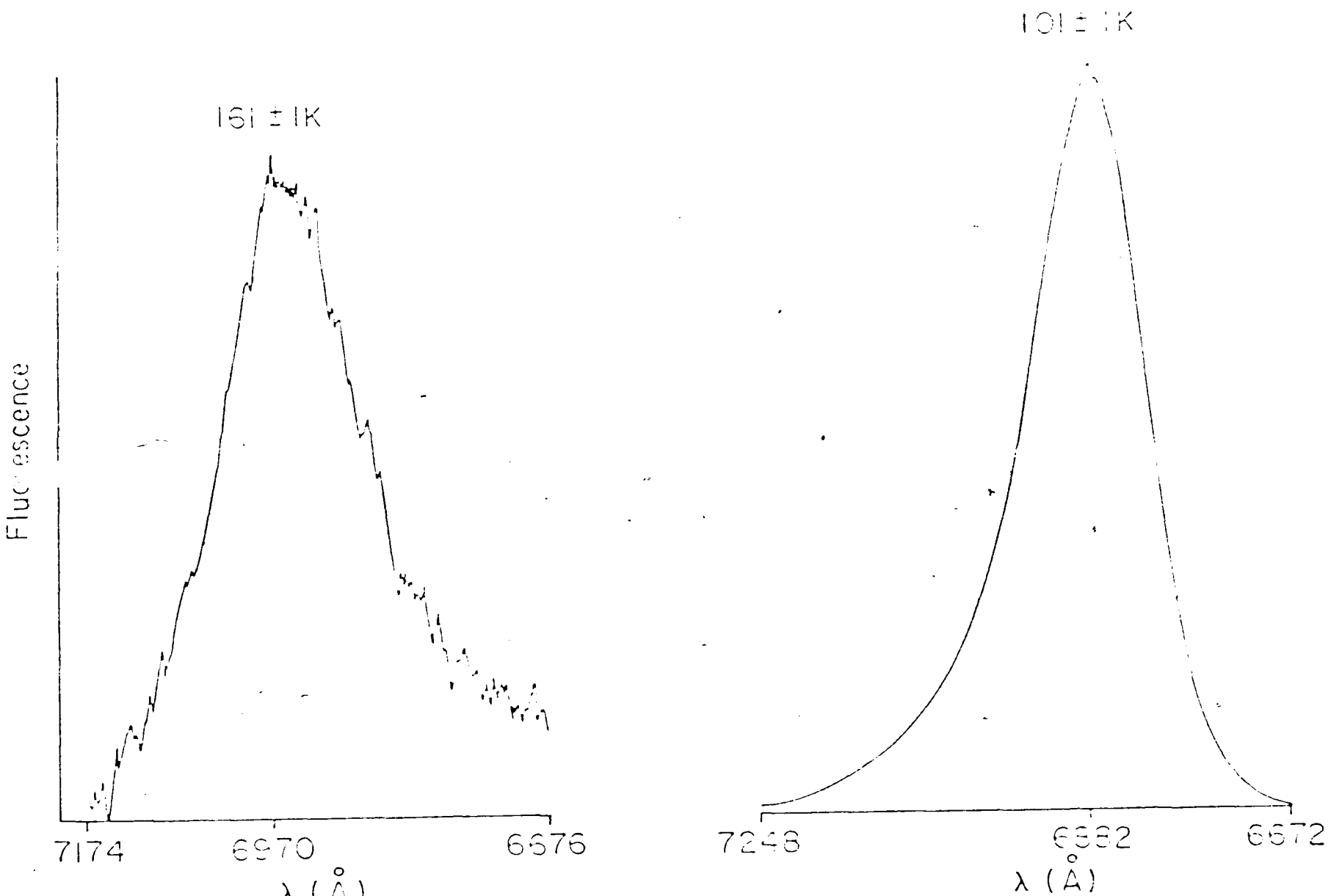
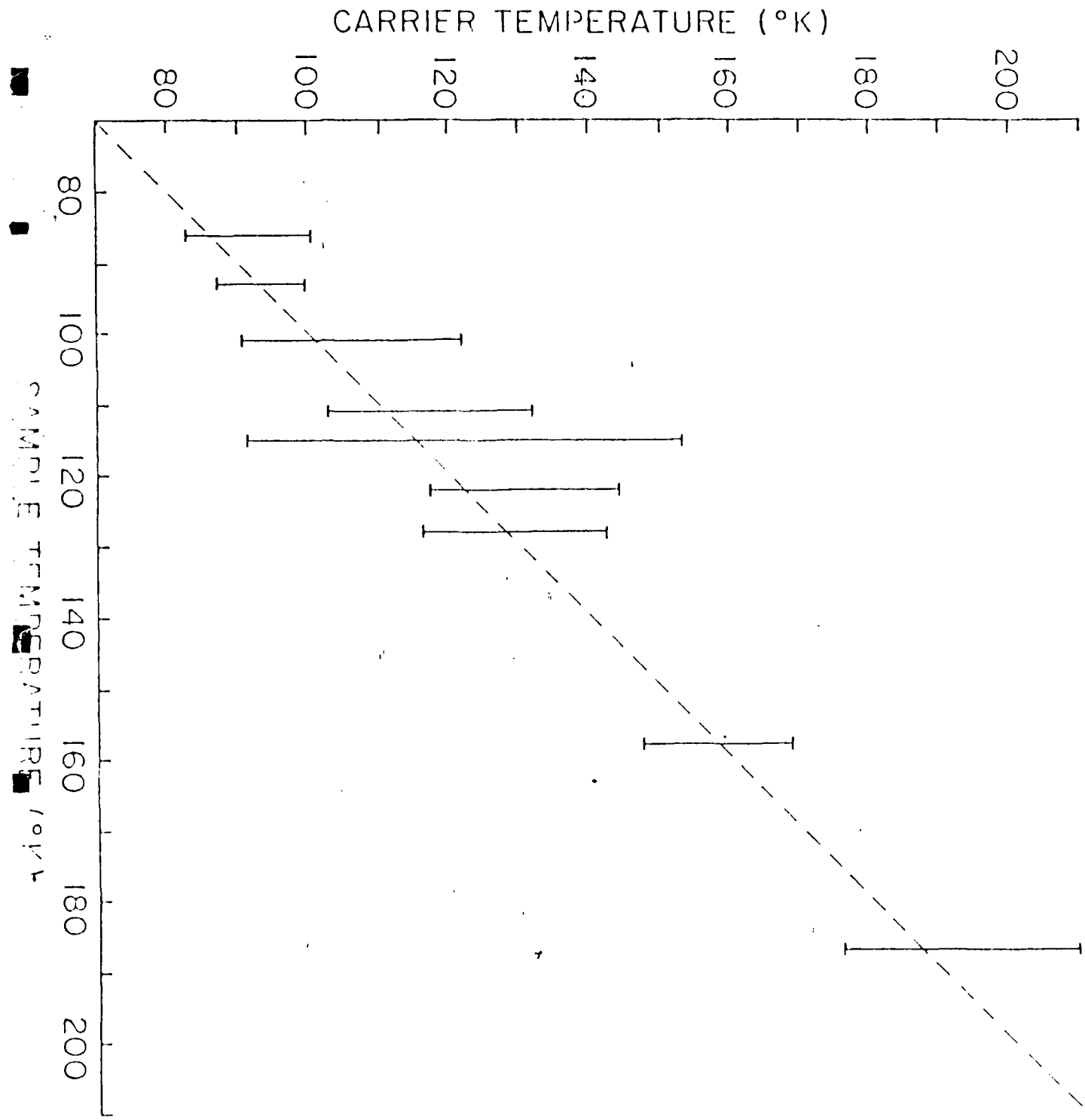


Figure 4


CARRIER TEMPERATURE (°K)
80
100
120
140
160
180
200

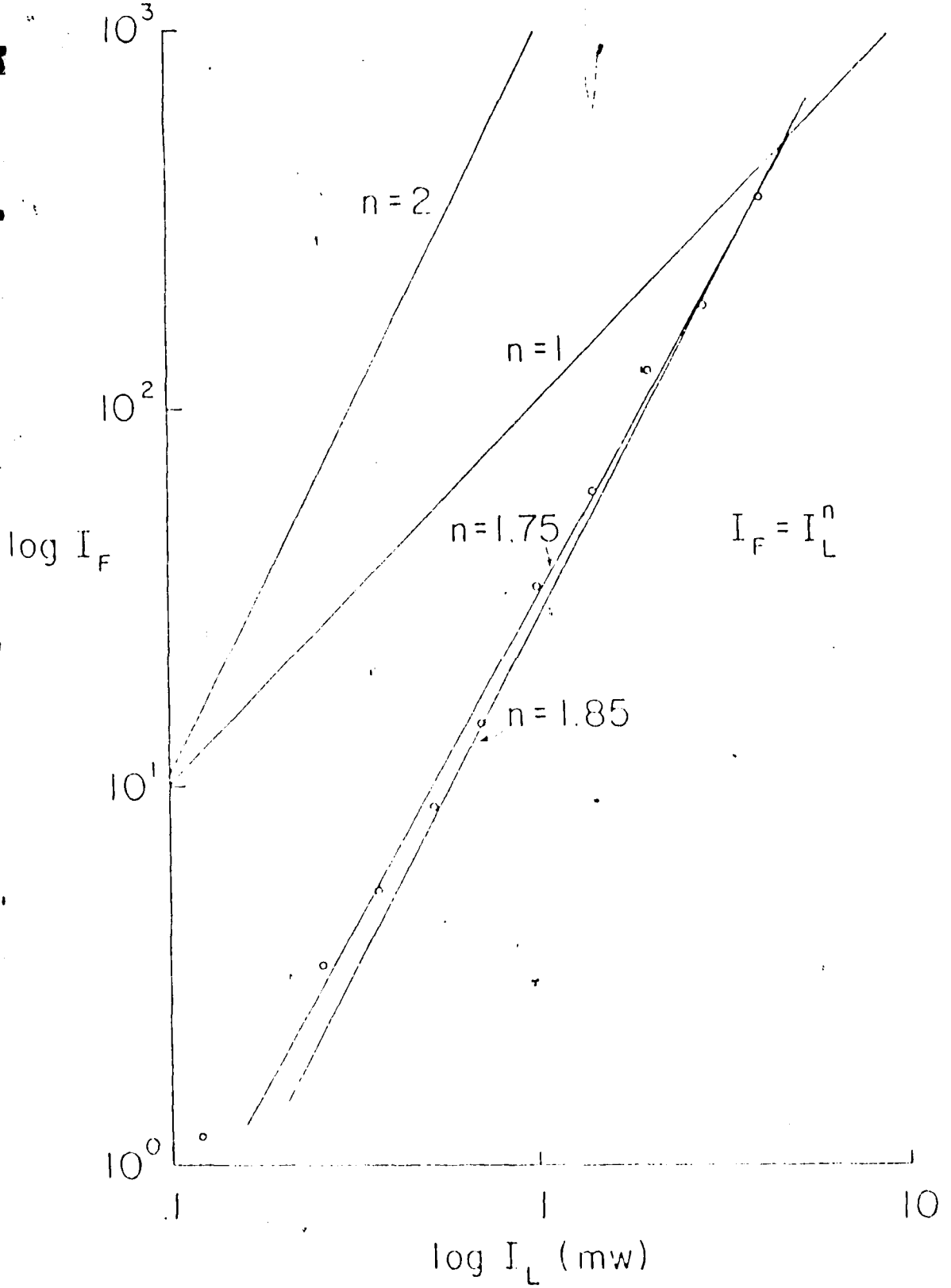


Figure 6

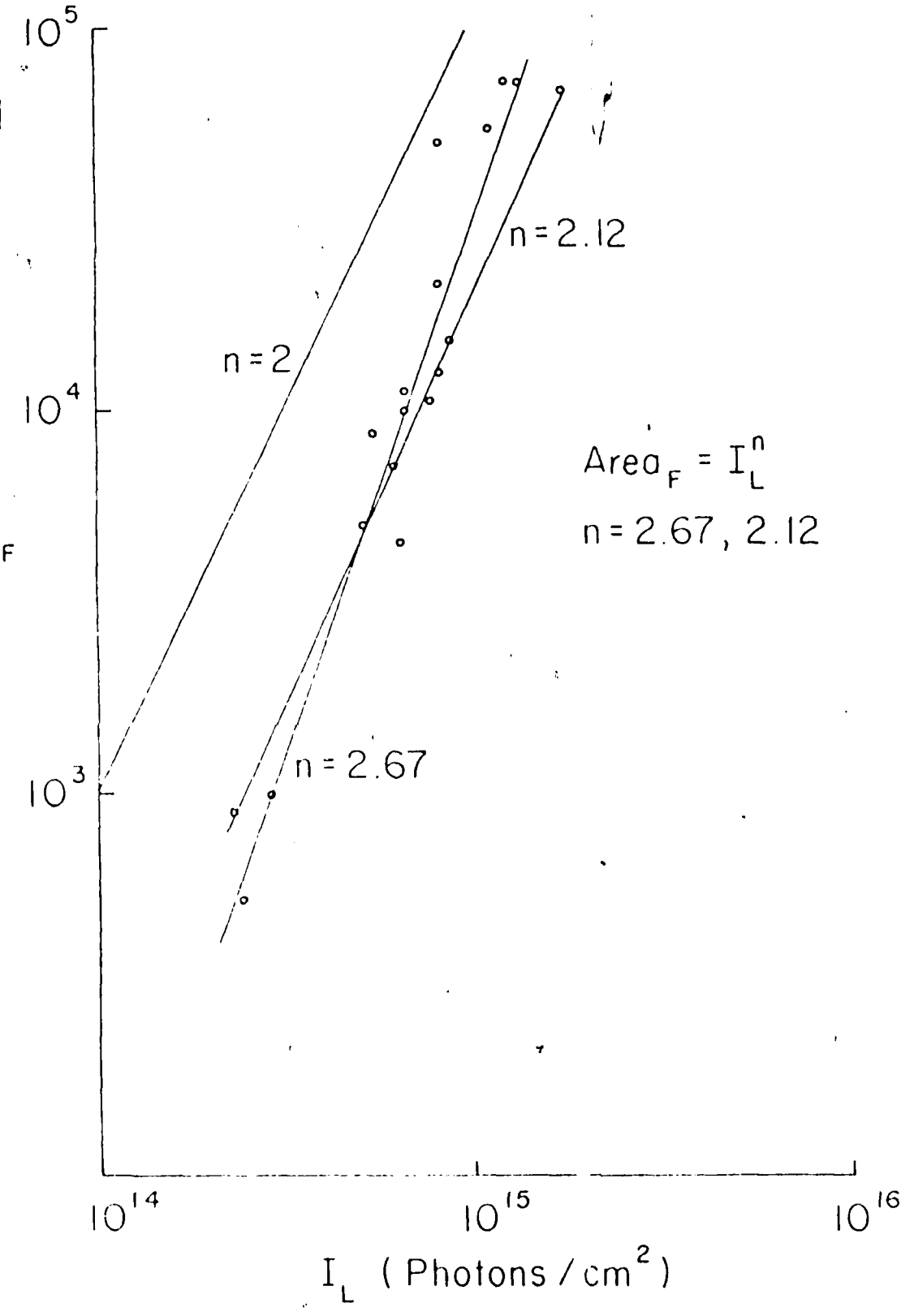

10^5
10^4
10^3
F
n = 2
n = 2.12
n = 2.67
$Area_F = I_L^n$
n = 2.67, 2.12
10^{14}
10^{15}
10^{16}
I_L (Photons / cm^2)

Figure 7

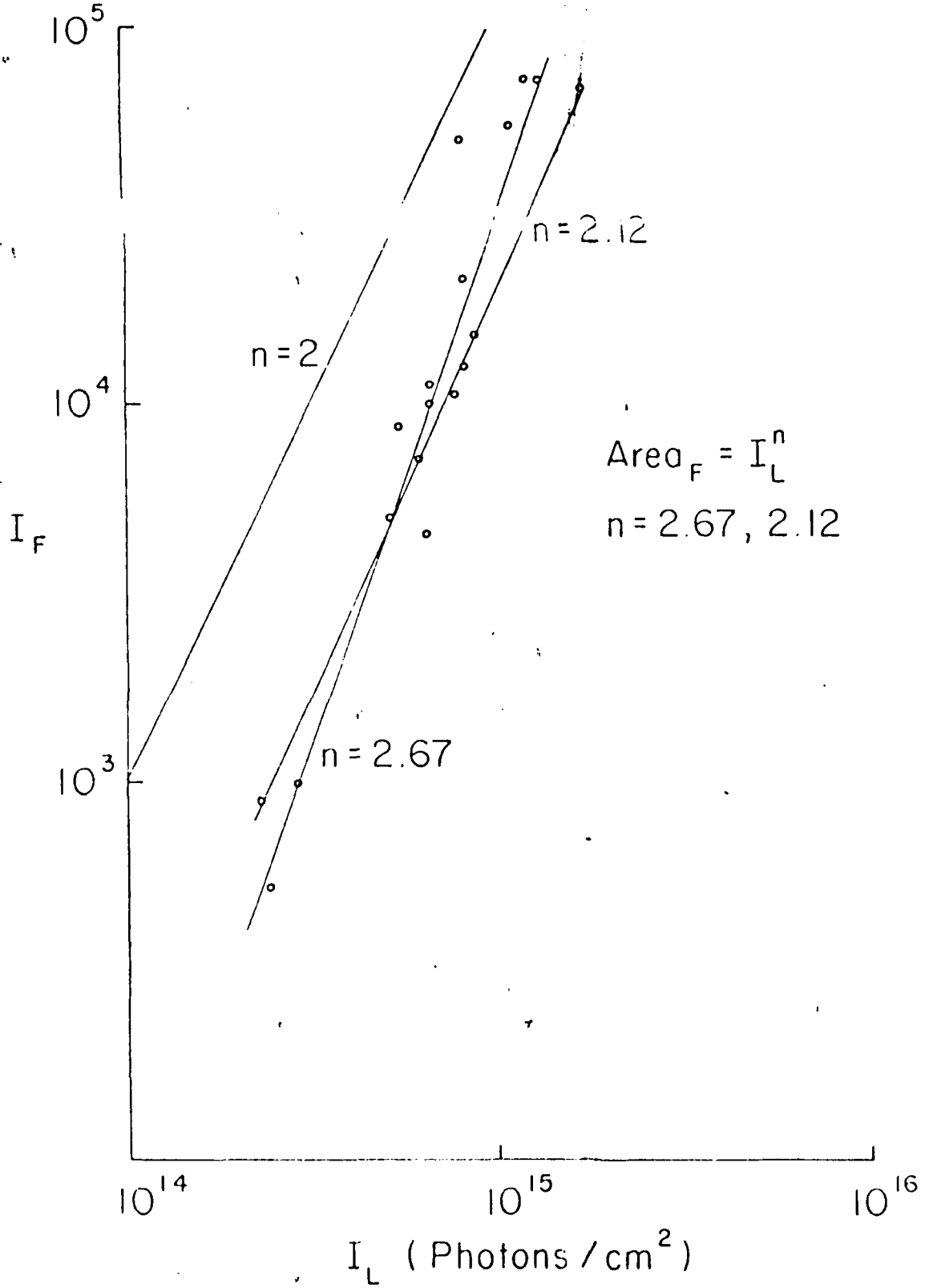
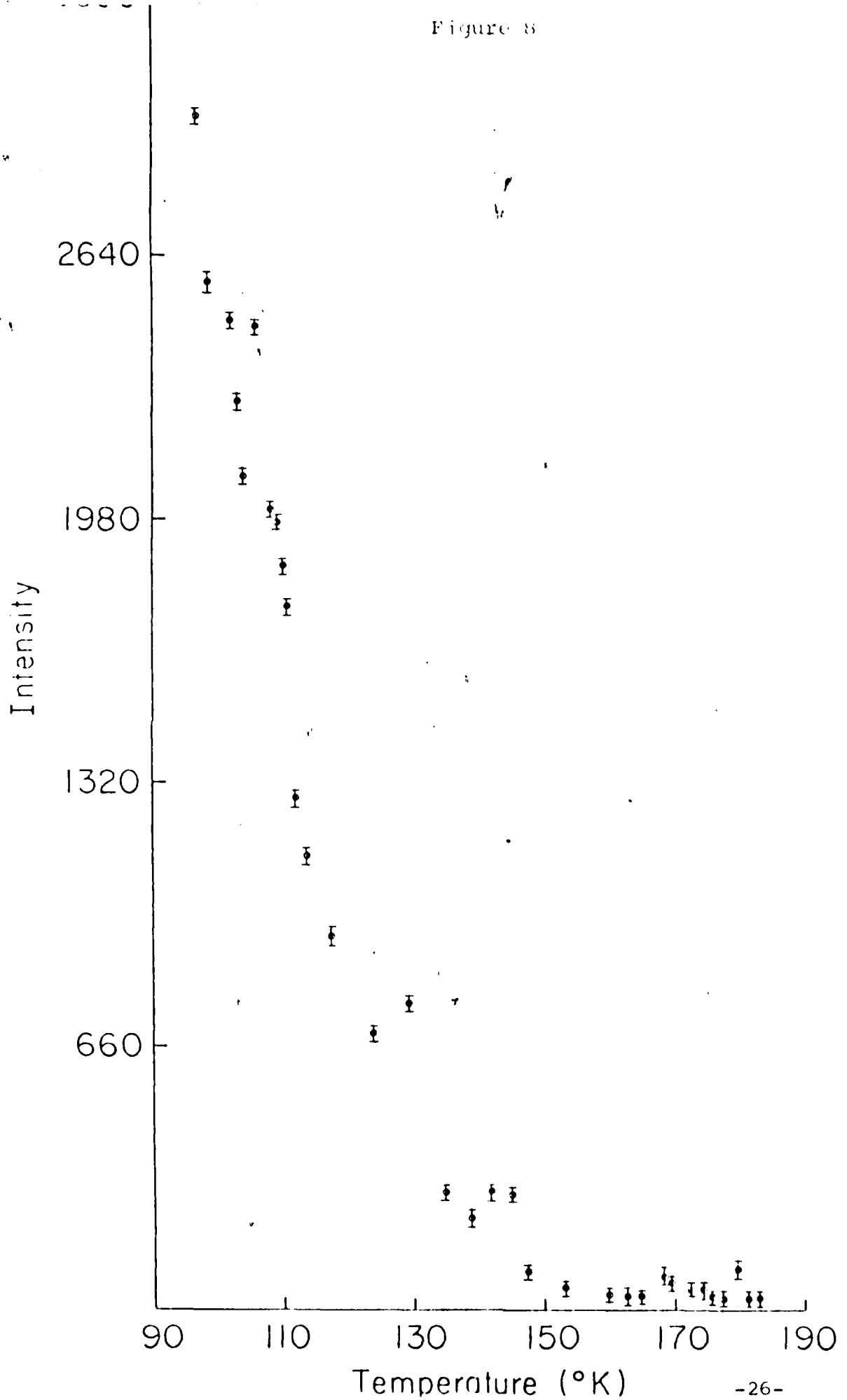

10^5
10^4
10^3
I_F
n = 2
n = 2.12
n = 2.67
$Area_F = I_L^n$
n = 2.67, 2.12
10^{14}
10^{15}
10^{16}
I_L (Photons / cm^2)

Figure 7

Figure 8


2640
1980
1320
660
Intensity
90
110
130
150
170
190
Temperature (°K)

B. GaAs

We have conducted a series of experiments on the fast emission kinetics of GaAs using intense picosecond excitation far above the band-gap. We have monitored the front surface luminescence as a function of time using a streak camera setup similar to the setup 1 on page 12. Different spectral regions were selected using narrow band interference filters. We observed the following:

1. For luminescent wavelengths longer than 7800 A we observed that the emitted light coincided in time with the excitation pulse.

2. For luminescent wavelengths shorter than 7800 A the luminescence persisted in time after the excitation pulse and the peak intensity occurred after the excitation.

3. When all wavelengths $\gtrsim$ 8200 A (to 9000 A, the limit of the streak camera spectral response) were monitored the delay in the luminescence peak was dependent on the excitation intensity.

We have shown the following preliminary conclusions. For the strongest excitations the absorption is strongly bleached resulting in a distribution of carriers much deeper into the sample than $1/\alpha$, the linear absorbtion coefficient. The delayed rise of the luminescence suggest that either more carriers are in the (100) and (111) valleys than the (000) (where the carriers are created and "slowly" transfer to the (000), or that the created electrons are very hot and the delayed rise is the cooling of these electrons. The lack of prolonged

emission wavelengths < 7800 A but less than the energy of the (100) and (111) valleys suggest that it is the second reason that causes this delayed rise.

2. SUBPICOSECOND LASER SYSTEM

A subpicosecond dye laser facility is under development at the Picosecond Laser and Spectroscopy Laboratory, City College of New York. The facility will include a fluorescence, an absorption and a Raman spectroscopic setup. It will be routinely used to measure ultrafast processes in semiconductors and other systems.

The following is a summary list of the progress achieved:

1. CW dye laser was built.
2. Different cavity configurations are being tested.
3. Experiments with the parameters of the laser (length of the cavity, dye concentrations, relative positions of the elements, e.g.) are being carried out to achieve optimum conditions.
4. Work on the long-term stability is being done.
5. Autocorrelation apparatus has been built to measure pulse width.
6. The laser reliably produces pulses of 2 psec duration with DODCI as a mode-locking dye in combination with malachite green.
7. The laser amplification system is being planned.

Considerable progress has been made with respect to the construction and characterization of the dye femtosecond laser. A number of different configurations have been tested. The two cavity designs that were studied are shown in figures 1 and 2. In the cavity (fig. 1) the lasing dye (Rh-6-G) and the mode-locking dye are flown together. The advantage of this design, easy alignment is

outweighed by the instability caused by the photodegradation of the mode-locking dye. Consequently, most of our effort has been focused on the second design following the design by Ippen and Shank . This cavity design is shown in figure 2. A number of variations have been tried such as the different cavity lengths, the elimination of the prism and more recently, we plan to substitute the focusing lens with a focusing mirror (mirror D in fig. 2).

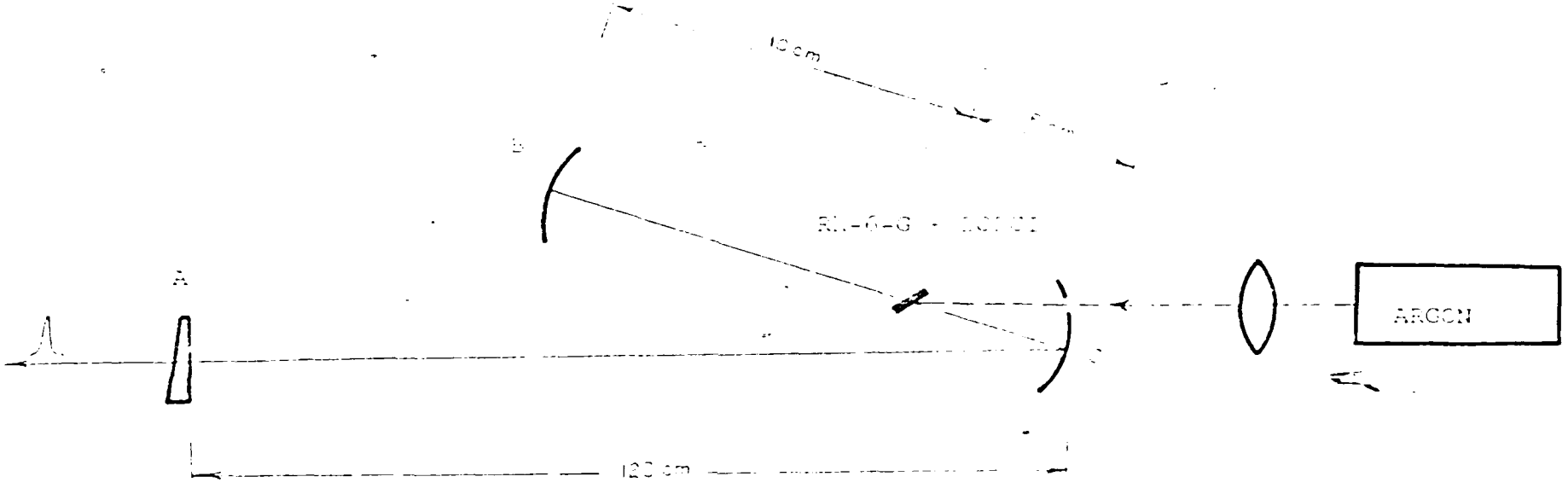
The effort to improve the long-term stability of the laser has led into the study of dye deterioration especially of the mode-locking dye DODCI. A copy of this manuscript is attached at the end of this section. Consequently, the stability has been improved by redesigning the flow systems for the dyes. Other sources of instability has been found to be air currents and dust and have been eliminated by enclosing completely the cavity.

Figure 3 shows the autocorrelation of the pulse. From 3a the pulse width is estimated to be 5.6 psec. The spike on top indicates a subpicosecond pulse riding on top of the broad 5.6 picosecond pulse. Figure 3b shows a 3.6 psec pulse, which is more typical to the current state of the dye laser without malachite green. It should be noted that in both cases only DODCI was used as a mode-locking dye. Combining DODCI and malachite green results in a autocorrelation profile shown in fig. 4. The pulse duration is typically $\sim$ 2 ps.

Our energies in the next four months will be directed towards decreasing the pulse width to below a picosecond, gaining a better insight of the parameters affecting the stability of the

laser and installing the grating pair for pulse compression to 500 fs and start building the amplification stage.

Figure 5 shows the new design of the laser oscillator currently under construction. Figure 5a is an overlay drawing of the cavity optics. Figure 5b is the actual technical drawing of the optical cavity mounts. These drawings can be used to build a mode-locked dye laser.



A. 99% Flat mirror

B.& C. 100% Rad cm.

Figure 1

Optical components

P.R=6cm. HR Coating 58[illegible]-630nm.
R.Brewster Angle Prism (SF)C0044-000
S.R=1 cm. HR Coating 500-630nm.
T.R=1 cm. HR Coating 58[illegible]-630nm.
X.Laser Model 99 [illegible]rivity at 610

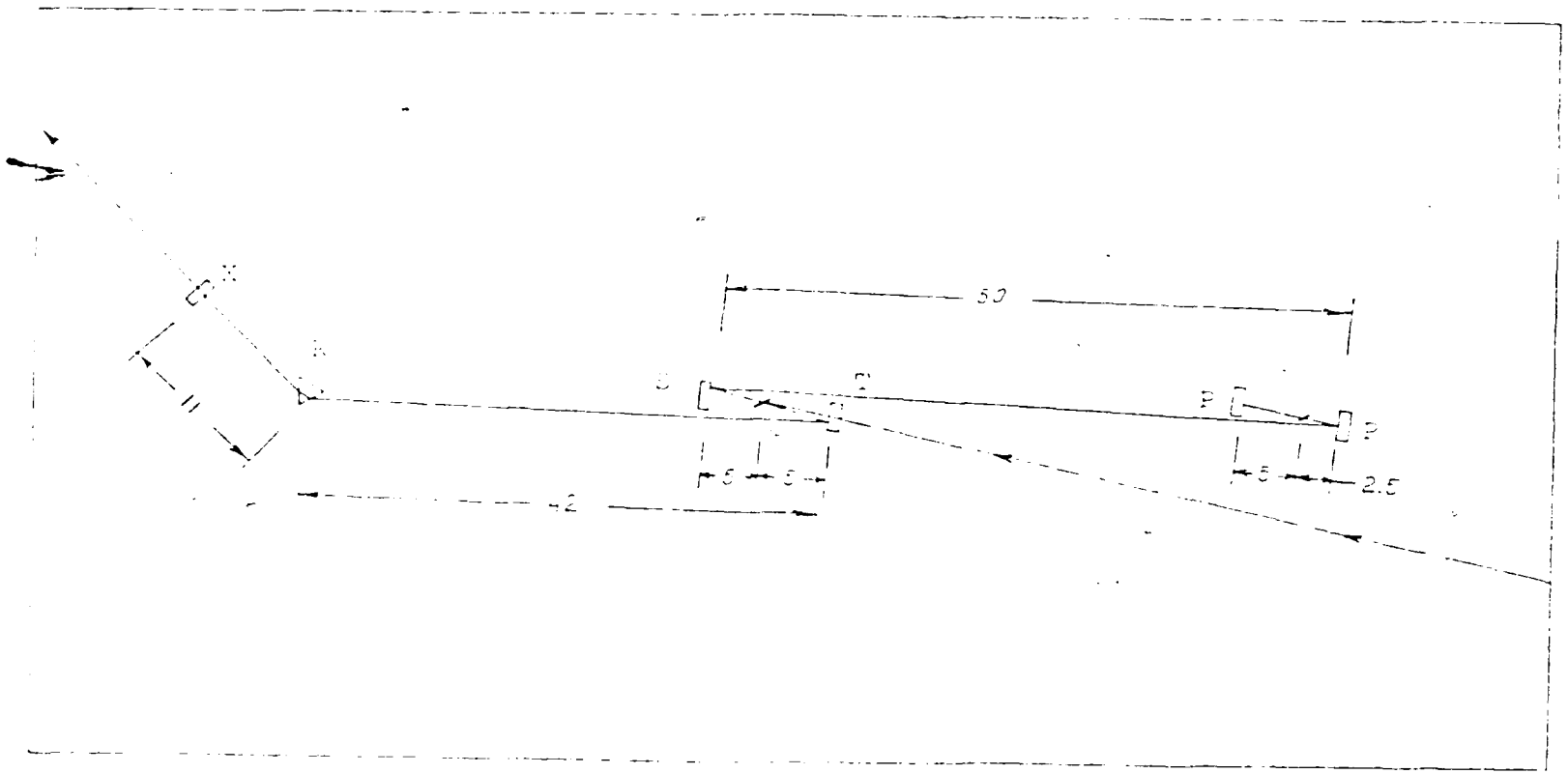


Figure 2a

Mechanical components

H.Mirror mounts (Linos)
I.Prism table-POMO (Klinger)
J.Translation stages (Linos)
K.Micropositioner mod.-A (Line tool)
L.Nozzles part[illegible]-324-02 (Current Radiation)

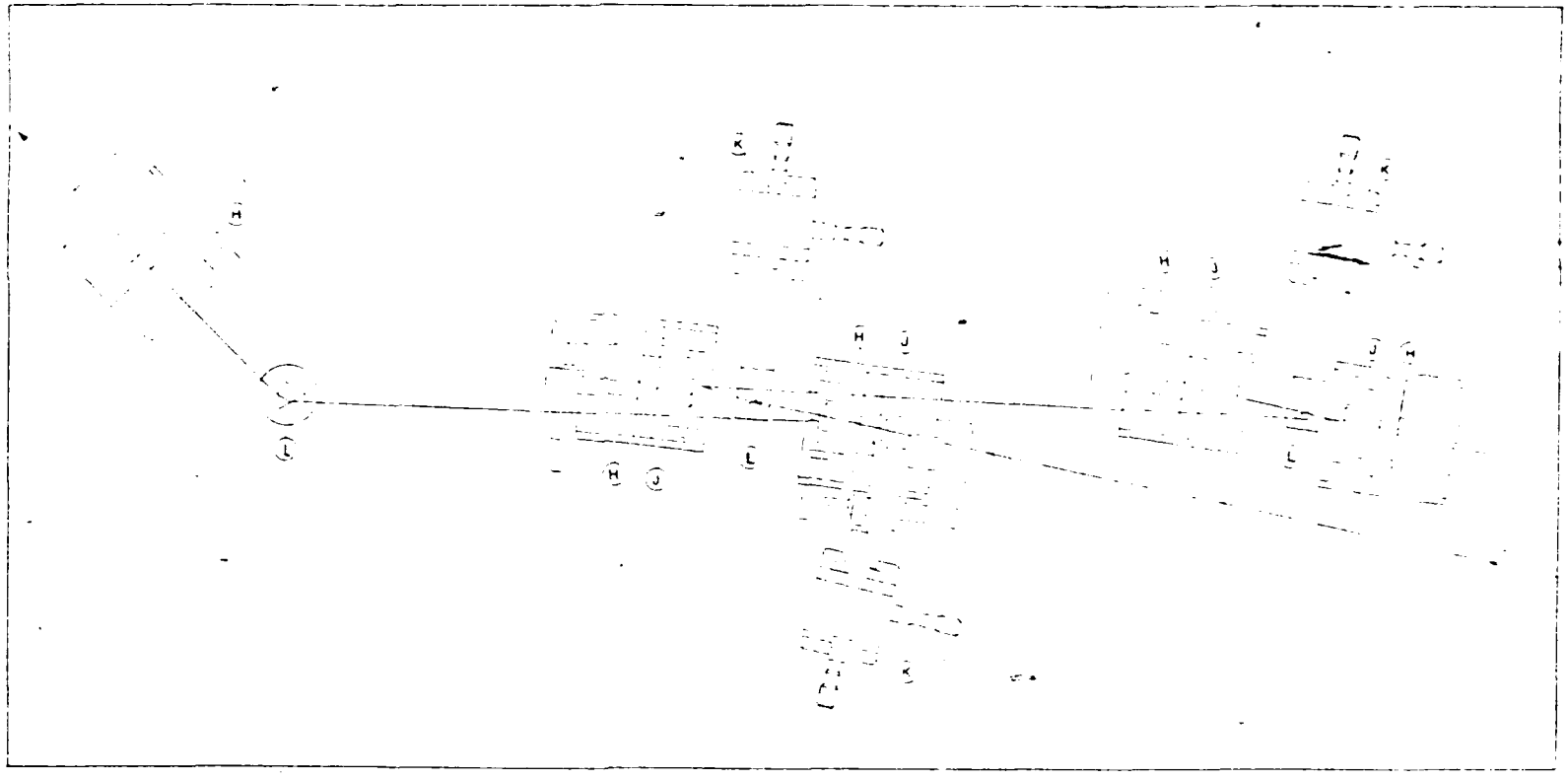
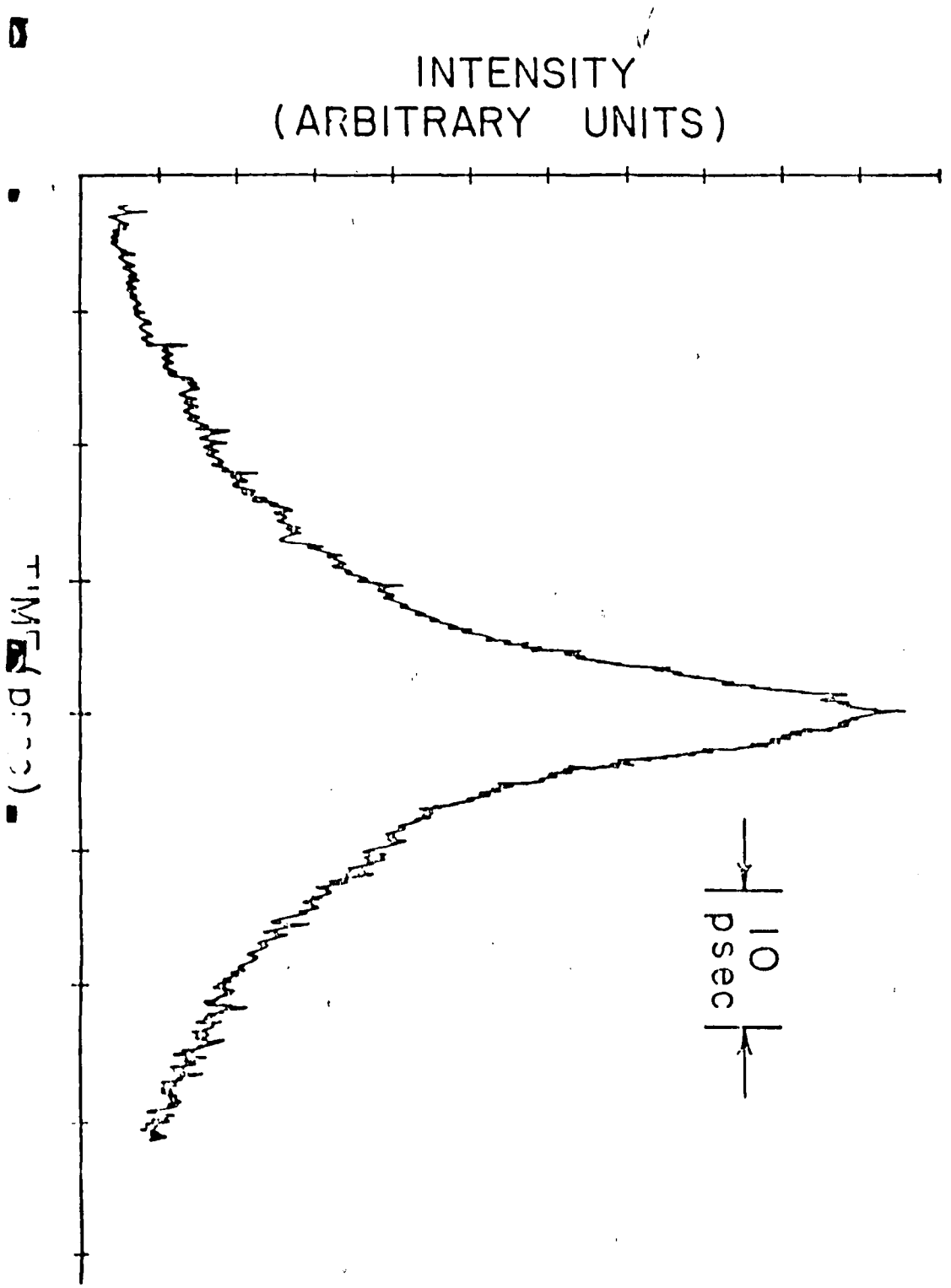


Figure 2b


INTENSITY
(ARBITRARY UNITS)
10 psec

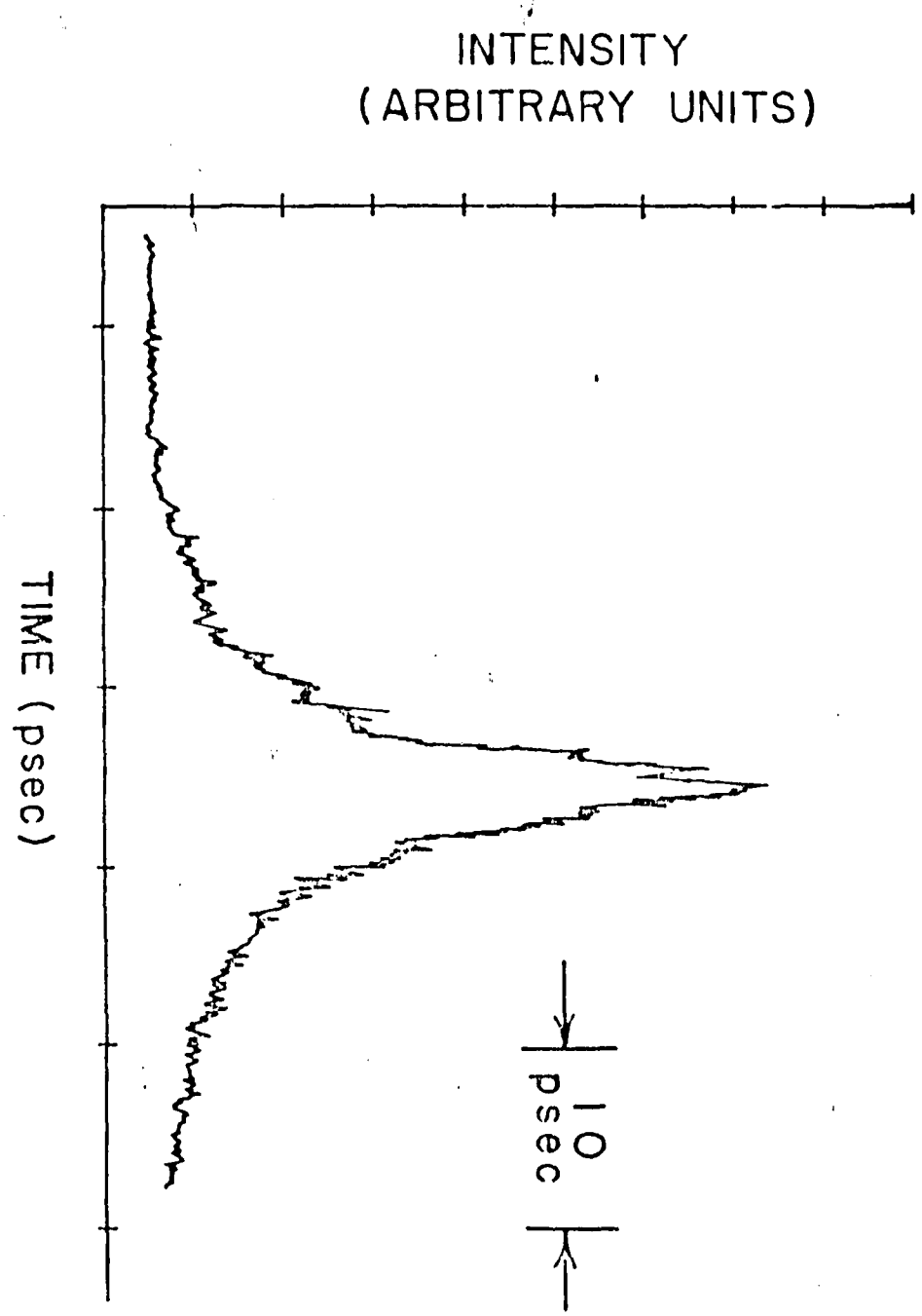


Figure 3b

Figure 4

Optical components

P.R=5cm. HR Coating 58[illegible]nm.
Q.R=∞cm. HR Coating 58[illegible]30nm.
R.Brewster Angle Prism (S-F)G[illegible]44-000
S.R=10cm. HR Coating 5[illegible]-63[illegible]nm.
T.R=10cm. HR Coating 58[illegible]-630nm.
U.R=20cm. HR Coating 54[illegible]-64[illegible]nm.(S-F)33836-[illegible]
V.R=10cm. HR Coating 54[illegible]-64[illegible]nm.(S-F)33837-[illegible]
W.R=∞Aluminum (NRC

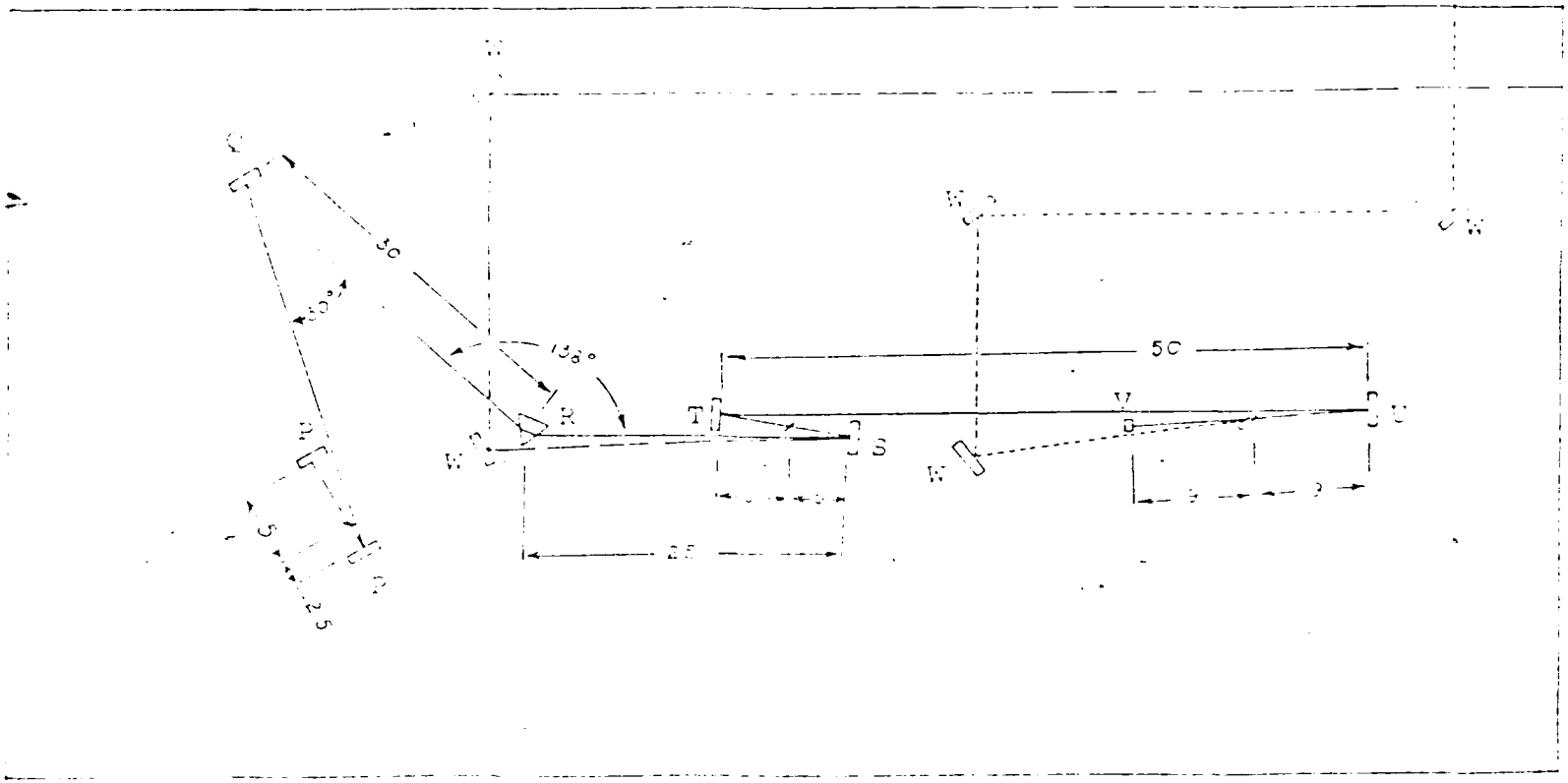


Figure 5a

Mechanical components

A.Mirror mounts-SL50 (Klinger)
B.Translation stages (Lansing)
C.Mirror mounts -SL25.4 (Klinger)
D.Mirror mounts-MM2 (NRC)
E.Mikropositioner mod.-A (Line tool)
F.Prism table-P046 (Klinger)
G.Nozzles pat#406-224-02 (Coherent radiation)

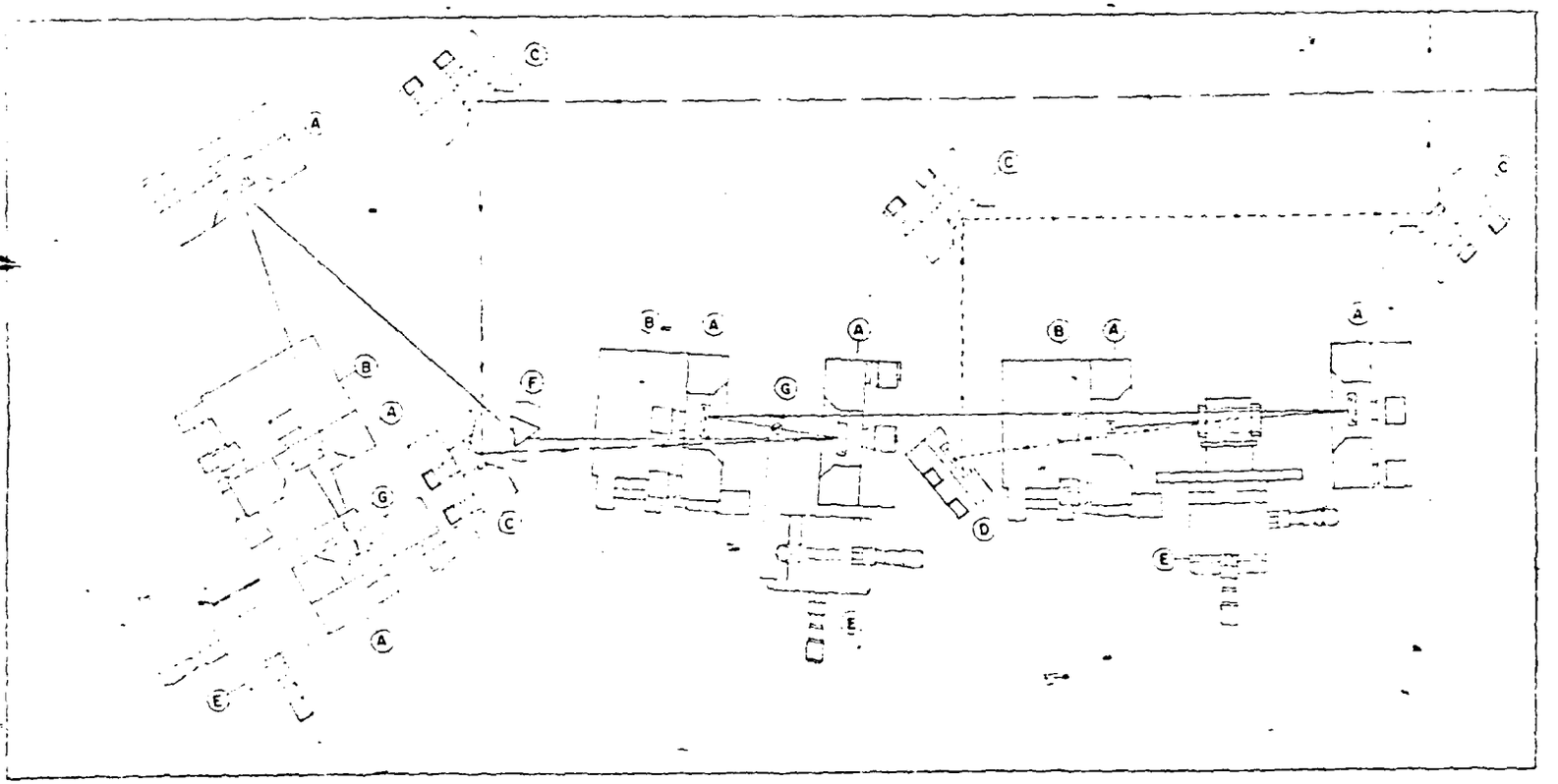


Figure 5b

Figure 6 shows a schematic design of the amplification stage. The Nd:Yag laser and the second harmonic crystal have already been tested. They will be used to pump the three cells containing Rh. This stage is expected to amplify the pulse 10^6 to 1 mj or to a power level of $\sim$ 2GW. The parts will be ordered before this grant starts.

The laser is currently operating at 6100 A. The wavelength of the amplified pulse can be shifted to other wavelengths by such non-linear techniques as SHG and SRS. In addition, focusing the beam of CCl_4 can produce by self-phase modulation and four photon mixing a broad-band known as the "continuum" which covers the entire visible spectrum.

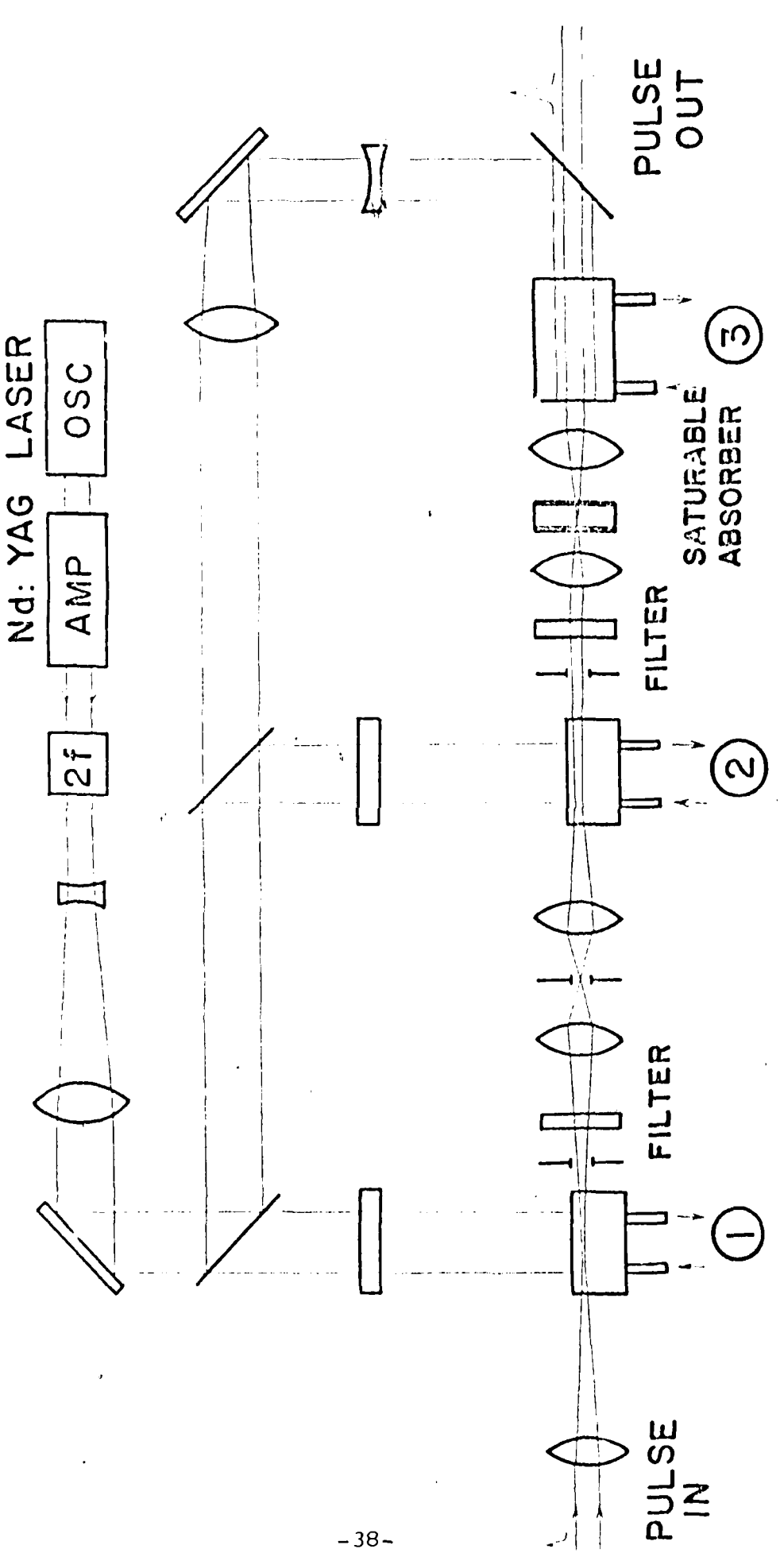

Nd: YAG LASER
OSC
AMP
2f
PULSE IN
FILTER
FILTER
SATURABLE ABSORBER
PULSE OUT
1
2
3

Figure 6

V. SECOND YEAR PROGRESS REPORT

This report summarizes the progress achieved during the second year in the research effort supported by AFOSR grant 80-0079. This section is divided into four parts, describing the highlights of the following four projects:

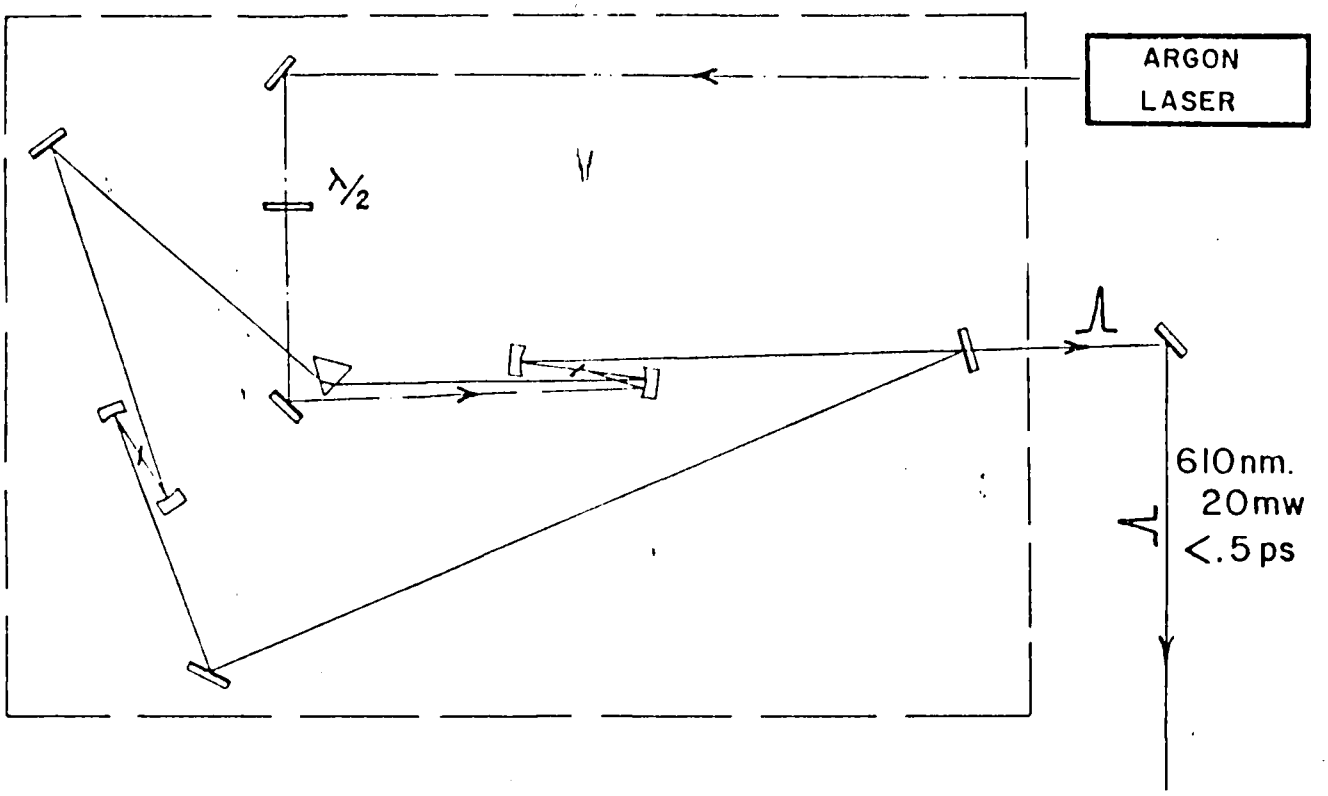
1) Development of the femtosecond laser instrumentation.
2) Luminescence measurements in semiconductors by novel subpicosecond population mixing technique.
3) Relaxation of photoexcited hot carriers in semiconductors by picosecond time resolved spectroscopy.
4) Luminescence from ferromagnetic and layered semiconductors by picosecond time resolved spectroscopy.

1) Femtosecond Laser Instrumentation

A reliable CW mode-locked ring oscillator has been completed during the last year. We will summarize here the operation and design of the final cavity design. A previous progress report dated 8/25/80 described in detail the various other laser cavity designs tested.

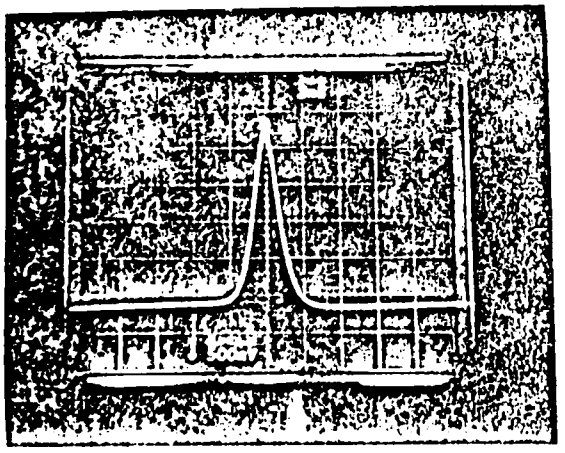
Figures 1 and 2 show the design and a photograph of the ring oscillator, respectively. The ring-cavity design has been found the most stable. The laser operates at 610 nm and consistently produces 0.3-0.4 psec pulses bandwidth limited on a daily basis. The laser is also tunable over a range, typically $\sim$200Å. A further major advance has been achieved in lowering the threshold pumping power from $>$3 watts to 0.9 watts. This has been accomplished by placing a $\lambda/2$ plate in the path of the pump Argon laser beam to change the polarization from "S" to "P". The expected overall improvement should be of the order of 15% not 300%. This indicates a more

MODE-LOCKED RING DYE LASER

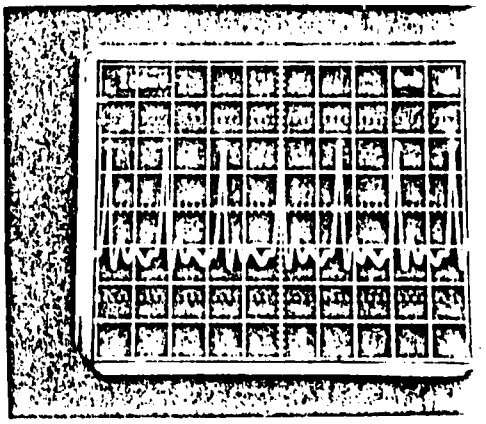


SHG CORRELATION

MODE - LOCKED TR

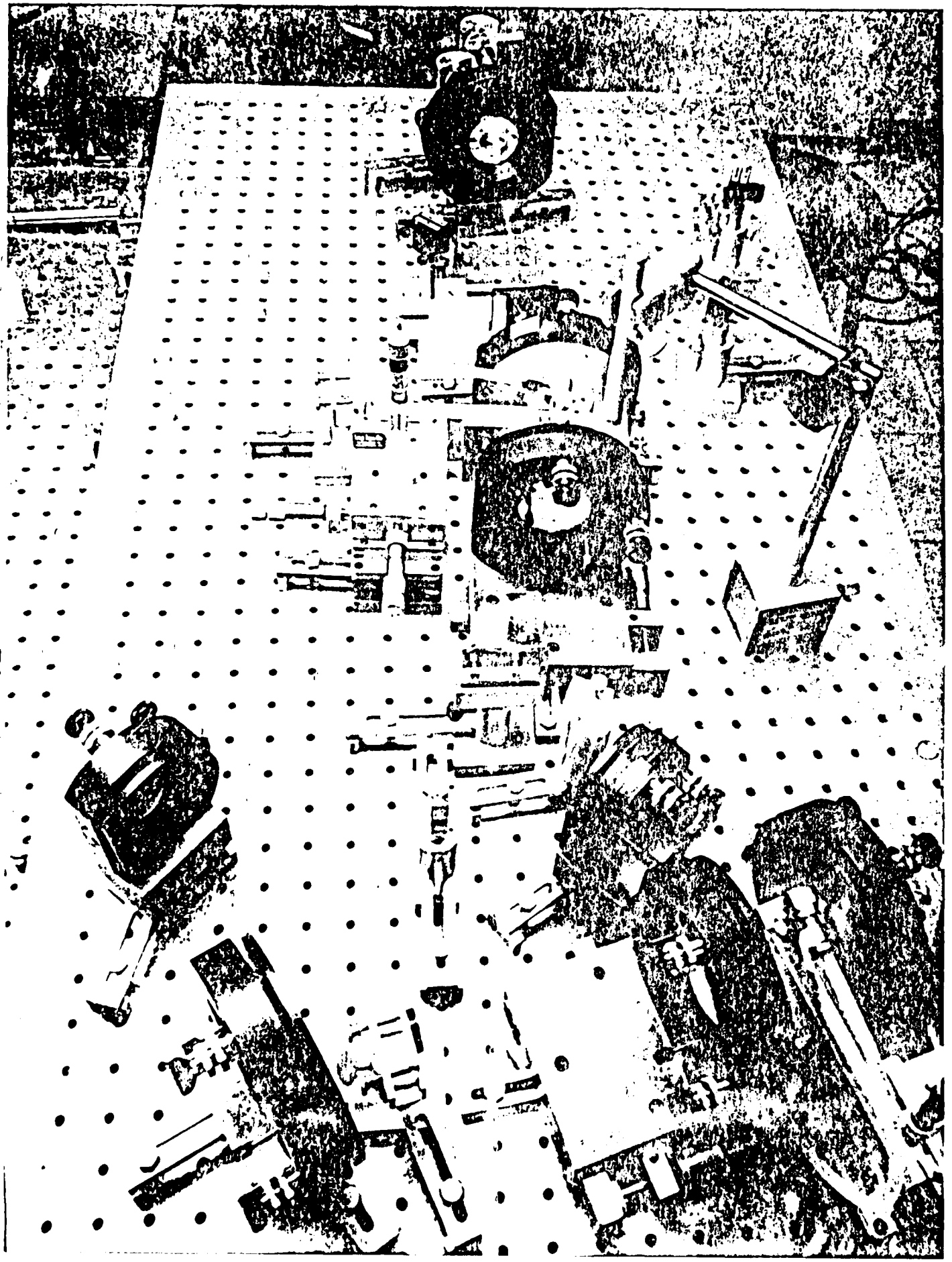


$\tau_p = .4$ ps



.6ps

Fig. 1



significant effect in the operation of these laser systems. These improvements have allowed us to reduce the output coupler reflectivity thereby obtaining substantially more output power. This is a breakthrough in the operation of the femtosecond dye laser using less than a watt pump power. The combination of the novel subpicosecond population mixing technique (see section II) and the improvements in the operation of the dye laser will allow scientists to use a less expensive subpicosecond fluorescence apparatus and pump Argon laser.

The subpicosecond pulses generated by the mode locked ring dye laser are of low energy, a few picojoules per pulse. In order to increase the energy of the pulses an amplifier is used. Fig. 3 shows the schematic design of the three dye stage amplifier. The amplifier stages are pumped by the second harmonic (530 nm) of a Nd:Yag laser. This system is under construction.

The important consideration in the amplifier is the synchronization of the Nd:Yag pulses with the pulses from the dye laser. The problem is not trivial. The repetition rate of the dye laser is 125 MHz while the Nd:Yag is variable from 0-10 pps. In addition the relative jitter between the two pulses should be as small as possible to improve the stability of the laser-amplifier system. Fig. 4 shows the schematic diagram of the electronics used to trigger the Nd:Yag laser. In addition, we have modified the Nd:Yag laser to decrease the inherent jitter from $>$ 2 nsec to $\sim$ 500 picoseconds. This is accomplished by disconnecting the internal pulse generator to pockell cell and instead connecting a Pulsar Associates model TWM-30 pulse generator. This pulse generator has a subnanosecond rise time and $<$ 300 picosecond jitter. A $\lambda/2$ plate

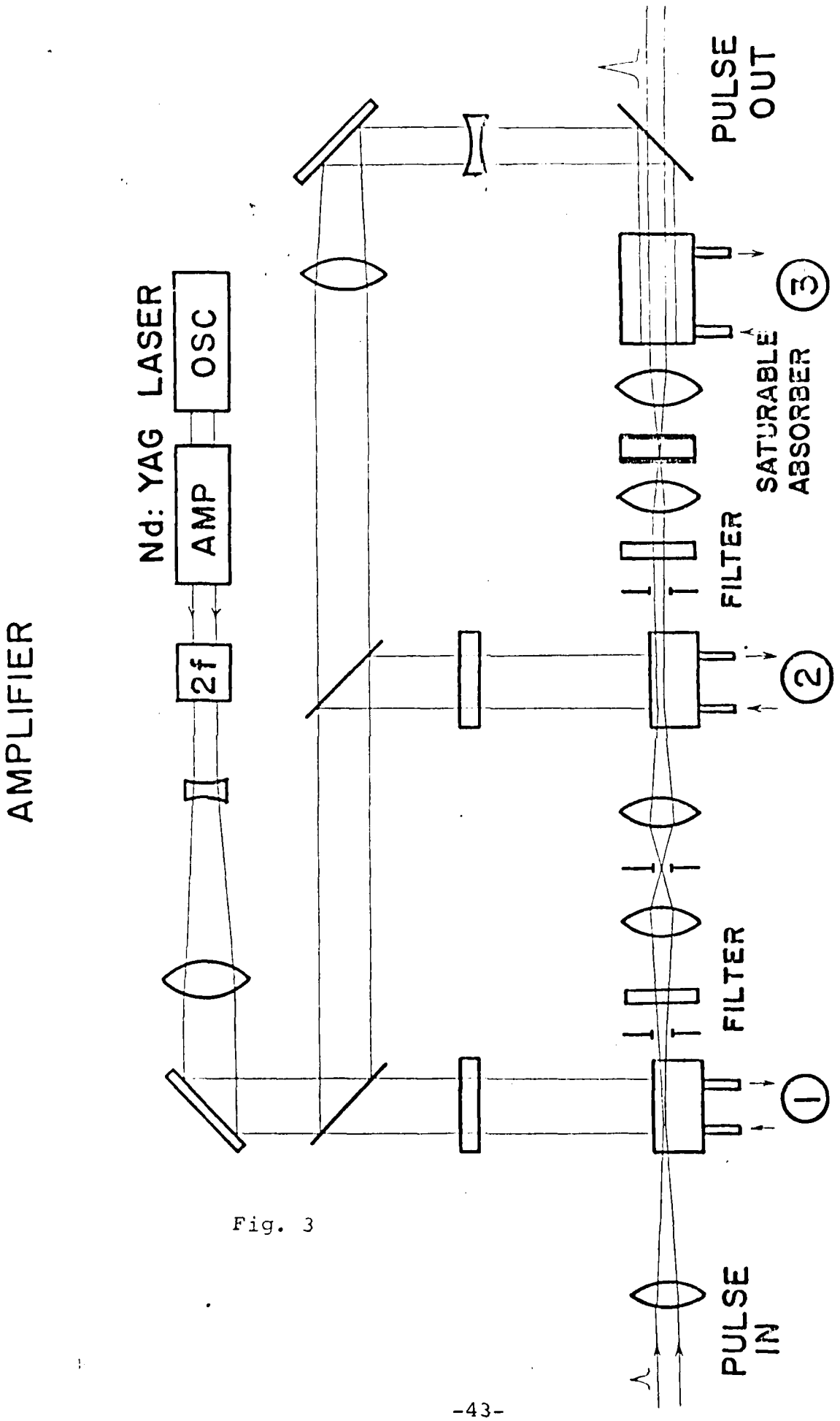

AMPLIFIER
Nd: YAG LASER
OSC
AMP
2f
PULSE IN
PULSE OUT
FILTER
FILTER
SATURABLE ABSORBER
1
2
3

Fig. 3

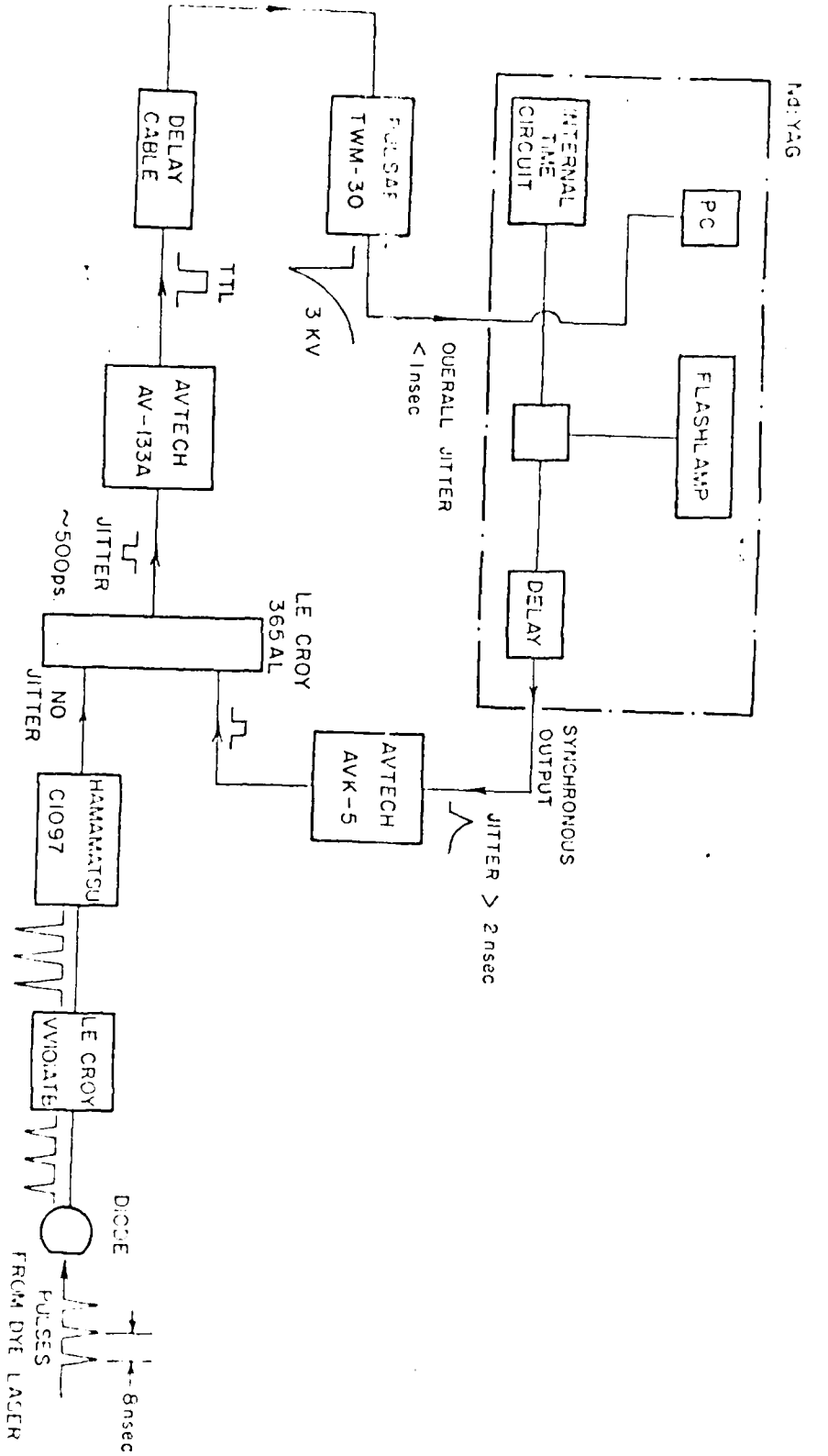

Nd: YAG
INTERNAL TIME CIRCUIT
PC
FLASHLAMP
DELAY
SYNCHRONOUS OUTPUT
JITTER > 2 nsec
OVERALL JITTER
< 1nsec
PULSAR TWM-30
3 KV
DELAY CABLE
TTL
AVTECH AV-133A
JITTER ~500ps
LE CROY 365AL
NO JITTER
AVTECH AVK-5
HAMAMATSU C1097
LE CROY
DIODE
PULSES FROM DYE LASER
8nsec

Fig. 4

also was inserted in the Nd:Yag cavity to prevent lasing action prior to the application of the high voltage pulse to pockell cell.

The synchronization of the Nd:Yag laser to the dye laser pulse is achieved by a coincidence technique. Pulses from the dye laser are detected by avalanche photodetector and amplified by a LeCroy (VV101ATB) pulse amplifier and fed to the LeCroy (365AL) pulse coincidence unit. On the other the hand, the synchronous output from the Nd:Yag laser triggers the Avtech pulse generator (AVK-V). The rise time of the pulses have a 100 picosecond rise time and the pulse width is adjustable. This is adjusted to be less than the period of the dye lasers, usually $\sim$ 6 nsec. When the two pulses coincide in the Lecroy 365AL the unit produces an output pulse. The output pulse is converted to a fast TTL by Avtech (AV-133A) which in turn triggers the Pulsar pulse generator that opens the pockell cell. There are two advantages in this design. First the system is immune to the jitter of the Nd:Yag electronics, since the pulse only opens the logic unit. The triggering occurs on the dye laser pulse which has practically no jitter. Second the electronics make use as much as possible of the internal electronics of the Nd:Yag laser with considerable savings in the overall cost.

The Hamamatsu delay unit is used to bring the two electronic pulses (the Nd:Yag and from the dye laser) to overlap at the coincidence unit. The overlap of the optical pulses at the first amplification stage is accomplished by adjusting the delay cable. The overlapping at the other two stages is insured because the path-length of the two beams are equal, see fig. 3.

2. Luminescence measurements in semiconductors by subpicosecond population mixing technique

A novel technique for time resolved luminescence spectroscopy has been developed. The technique shown in fig. 5 involves the irradiation of the sample by two subpicosecond laser beams modulated at different frequencies. The subpicosecond excitation pulses are delayed with respect to each other by a prism. The luminescence from the sample is conveniently detected by a slow photomultiplier and a lock-in amplifier at the heterodyne frequency; that is, the difference of the two modulating frequencies. No streak camera or fancy superfast electronics are required. The technique has been applied to p-type GaAs at 90K in order to study the ultrafast recombination of the photogenerated carriers. An analysis of the luminescence correlation function measured by the lock-in amplifier is shown in fig. 6. The part of the signal that is detected by the lock-in amplifier, is proportional to the product of the photogenerated holes and electrons produced by the two beams, that is:

$$I_s(\tau) \propto \int_{-\infty}^{\infty} n(t+\tau)\,p(t)\,dt + \int_{-\infty}^{\infty} n(t)\,p(t+\tau)\,dt$$

In these experiments, the excitation pulses are much shorter than the lifetimes of photogenerated electrons and holes. The expected form of the decay is two exponentials functions which contain the relaxation times of both the minority and majority carriers. The exponential fitting of the curve in fig. 6 gives a lifetime of 40 ± 10 psec in p-type GaAs at doping concentration of 6×10^{18} cm^{-3}. The measurements are in good agreement with streak camera measurements. It should be pointed out that the resolution

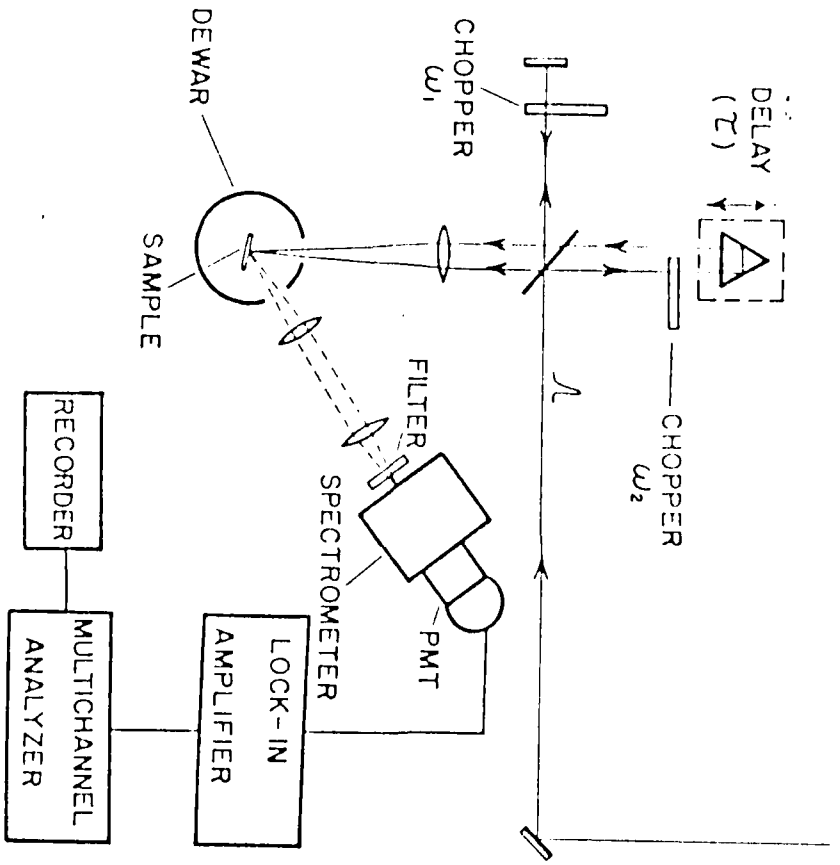

DELAY
(τ)
CHOPPER
ω1
DEWAR
SAMPLE
CHOPPER
ω2
FILTER
SPECTROMETER
PMT
RECORDER
MULTICHANNEL
ANALYZER
LOCK-IN
AMPLIFIER

Fig. 5

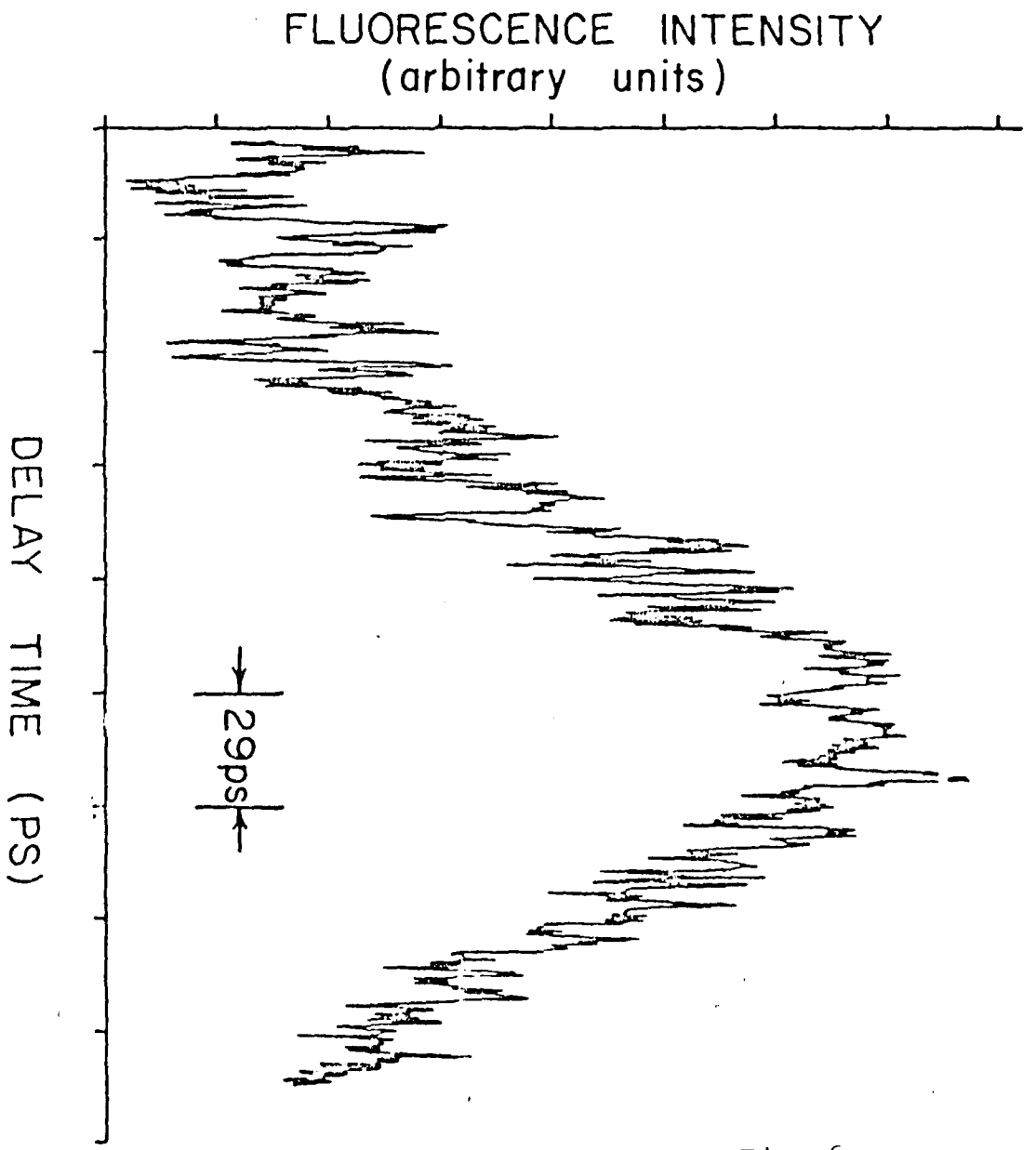

FLUORESCENCE INTENSITY
(arbitrary units)
DELAY TIME (PS)
29ps

Fig. 6

of the technique is only limited by the pulse duration of the laser pulse 0.4 ps. We plan to use this method to study CdSe.

3. Relaxation of photogenerated Hot carriers in semiconductors

Free carrier relaxation times in GaAs have been shown to increase as the free carrier density increases by intense picosecond photogeneration. Recently, R. J. Seymour, M. R. Junnarkar, and R. R. Alfano (see appendix) have measured the hot photoluminescence kinetics of GaAs under intense picosecond excitations (10^{28} photons/cm^2 sec). A slow risetime of the near bandedge luminescence has been observed arising from a slowed cooling of the electron distribution. The slowed kinetics can be attributed to the high optical phonon density and screening of the electron-phonon interaction. There are two theories which can explain the relaxation time of photogenerated hot electrons when the concentration of the hot carriers is very high in the sample GaAs. One is the screening of hot electrons on the emissions of LO-phonons from electrons. The other is the heated LO-phonon effect on the slowed relaxation time of the hot electrons. We have shown by comparing our experimental results to the two theories, that screening of the electron-phonon interaction is responsible for the observed slowing. These results are important in understanding the mechanisms of laser annealing. The effect of screening of e-phonon interaction by photon generated carriers is being investigated in other semiconductors i.e. GaSe (see section 4).

4. Luminescence from ferromagnetic and Layered semiconductors

We shall review the research accomplished on the magnetic semiconductor, $CdCr_2Se_4$. Research performed during the last period of this grant determined the location of the fundamental

energy gap of $CdCr_2Se_4$ at 1.8 ev. This has been disputed by many physicists for the last fifteen years. A manuscript was written and a paper was published in Physical Review Letters, 42, 558 (1981), which described our work in great detail.

Continuing this type of research, we found there is an abrupt and dramatic change in the photoluminescence intensity below the sample's Curie temperature 130K. This is a new phenomenon which is different from the observed in nonmagnetic semiconductors. We have attributed this effect to the ferromagnetism of $CdCr_2Se_4$. However, no theory has yet been developed. A paper is being written describing our observations.

A higher power picosecond laser was used to excite the sample $CdCr_2Se_4$. The emission band was found to broaden and shift to red as the excitation intensity increased. This emission was ascribed to the electron hole plasma effect.

We shall now review our research of layered semiconductors. GaSe is a layer type semiconductor with interesting optical properties of absorption and emission. One particular aspect about GaSe is that it may be an indirect gap semiconductor. Thus, the intervalley scattering rate can be found with the help of the time resolved absorption and emission spectroscopy. The dynamical processes of free carriers in this compound have not yet been determined. In order to understand the physics of the relaxation, the intervalley scattering rate and the decay of free carriers, we have initiated two types of experiments: the time resolved absorption spectroscopy measurement and the high excitation photoluminescence spectral measurements. The first type of experiments gives us the information on the relaxation time of hot

carriers. The second one confirms that there are two emission modes across the band gaps. We are currently working on these problems.

VI. THIRD YEAR PROGRESS REPORT

Significant progress has been achieved during the current year in the research areas supported by AFOSR. The following list highlights our achievements.

1. Achievements Summary During 1981-1982

1.1. 18 articles published and submitted for publication in professional journals (see list).

1.2. 7 conference presentations (3 invited, see list).

1.3. 1 patent disclosure (see list).

1.4. Completed a compact femtosecond time measuring correlator.

1.5. Completed development of a tunable reliable femtosecond ring laser oscillator - 300 fs, 200 A.

1.6. Major advance - improve subpicosecond laser design by lowering the laser threshold to $<$ 1 watt.

1.7. Major advance - invented a femtosecond luminescence technique for probing semiconductor processes called population mixing.

1.8. Completed the first generation femtosecond laser amplifier.

1.9. Within just 2 years of effort in the subpicosecond dye laser field, we have at a university achieved parity and in some areas surpassed such an established laboratory. We started with an empty facility.

1.10. So far we have trained 6 scientists (2 professors, 1 postdoc, 2 graduate students and a technician) in the art of subpicosecond ultrafast technology.

1.11. Scientific Advances -

i. For highly photoexcited semiconductors, we have measured by picosecond fluorescence spectroscopy the screening of optical phonons in GaAs and GaSe.

ii. For a highly photoexcited semiconductor, we have observed and measured by picosecond absorption spectroscopy the reduction in the optical phonon deformation potential in GaSe.

iii. For the magnetic semiconductor $CdCr_2Se_4$ we have measured and observed for the first time by photo-luminescence spectroscopy, the splitting of degeneracy of S-conduction band.

iv. We found the recombination of carriers on $CdCr_2Se_4$ is "super ultrafast" $<$ 2 ps at 200 K increasing to only 20 ps at 77 K. The ultrafast recombination time may allow for development of the fastest optical switch at room temperature.

2. Research Progress

The research effort at the USL is concentrated in four projects:

A. Development of femtosecond laser system.

B. Luminescence measurements by population mixing.

C. Luminescence studies of ferromagnetic and layered semiconductors.

D. Relaxation of photoexcited hot carriers in semiconductors by picosecond time-resolved spectroscopy.

The following describes the progress of each project:

A. Development of Femtosecond Laser System

A reliable femtosecond laser system has been completed. The system consists of a subpicosecond passively mode-locked ring dye laser and an amplifier system. Previous progress reports dated 1980 and 1981 summarize the development of the laser oscillator and describe in detail the final cavity design. We plan to decrease the pulse duration to 70 femtoseconds by using a thinner jet for the saturable absorber of the order of 20 μ. This will allow us to have the capability of higher temporal resolution for the study of kinetics in this time domain.

The first generation of the amplifier system has been completed, it consists of a three stage dye amplifier chain (fig. 1). The three dye cells are pumped by the second harmonic of a Nd:Yag laser. The dyes circulated in the cells are red kiton 620 in the first and rhodamine 640 in the last two. The amplification of the system is $10^6 - 10^7$, or $\sim$ 0.2 mJ energy per pulse. Focusing the pulses to a 1 mm diameter produces powers $>$ 10 GW/cm^2. We plan to continue working on optimizing the parameters during the current funding period. The two major improvements include: (1) the replacement of Nd:Yag mirrors and beam splitters which do not

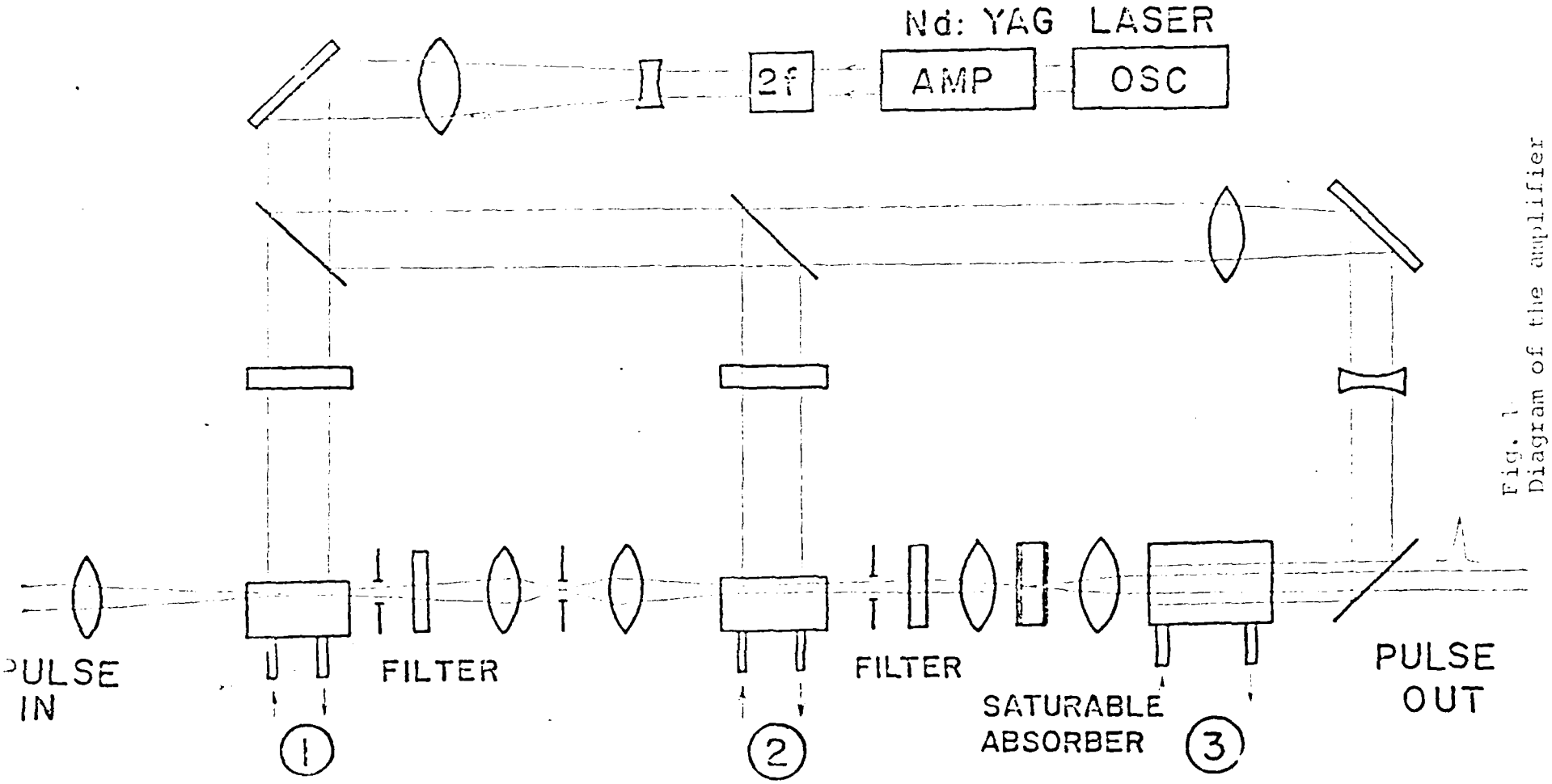


Fig. 1
Diagram of the amplifier

withstand the high power of the YAG laser. We have already contacted the CVI Corporation for coating specifications and we have already tested sample mirrors. The test has been successful. The sample mirrors do withstand the high power. (2) Add a fourth stage in the amplifier dye chain. We expect to increase amplification by another factor of 10. The fourth stage will also make more efficient use of the power of the Nd:Yag laser.

The important consideration in the amplifier is the synchronization of the Nd:Yag pulses with the pulses from the dye laser. The problem is not trivial. The repetition rate of the dye laser is 125 MHz while the Nd:Yag is variable from 0-10 pps. In addition the relative jitter between the two pulses should be as small as possible to improve the stability of the laser-amplifier system. Fig. 2 shows the schematic diagram of the electronics used to trigger the Nd:Yag laser.

The synchronization of the Nd:Yag laser to the dye laser pulse is achieved by a coincidence technique. Pulses from the dye laser are detected by avalanche photodetector and amplified by a LeCroy (VV101ATB) pulse amplifier and fed to the LeCroy (365AL), pulse coincidence unit. On the other hand, the synchronous output from the Nd:Yag laser triggers the Avtech pulse generator (AVK-V). The rise time of the pulses have a 100 picosecond rise time and the pulse width is adjustable. This is adjusted to be less than the period of the dye lasers, usually $\sim$ 6 nsec. When the two pulses coincide in the LeCroy 365AL the unit produces an output pulse. The output pulse is converted to a fast TTL by Avtech (AV-133A) which in turn triggers the Pulsar pulse generator that opens the pockell

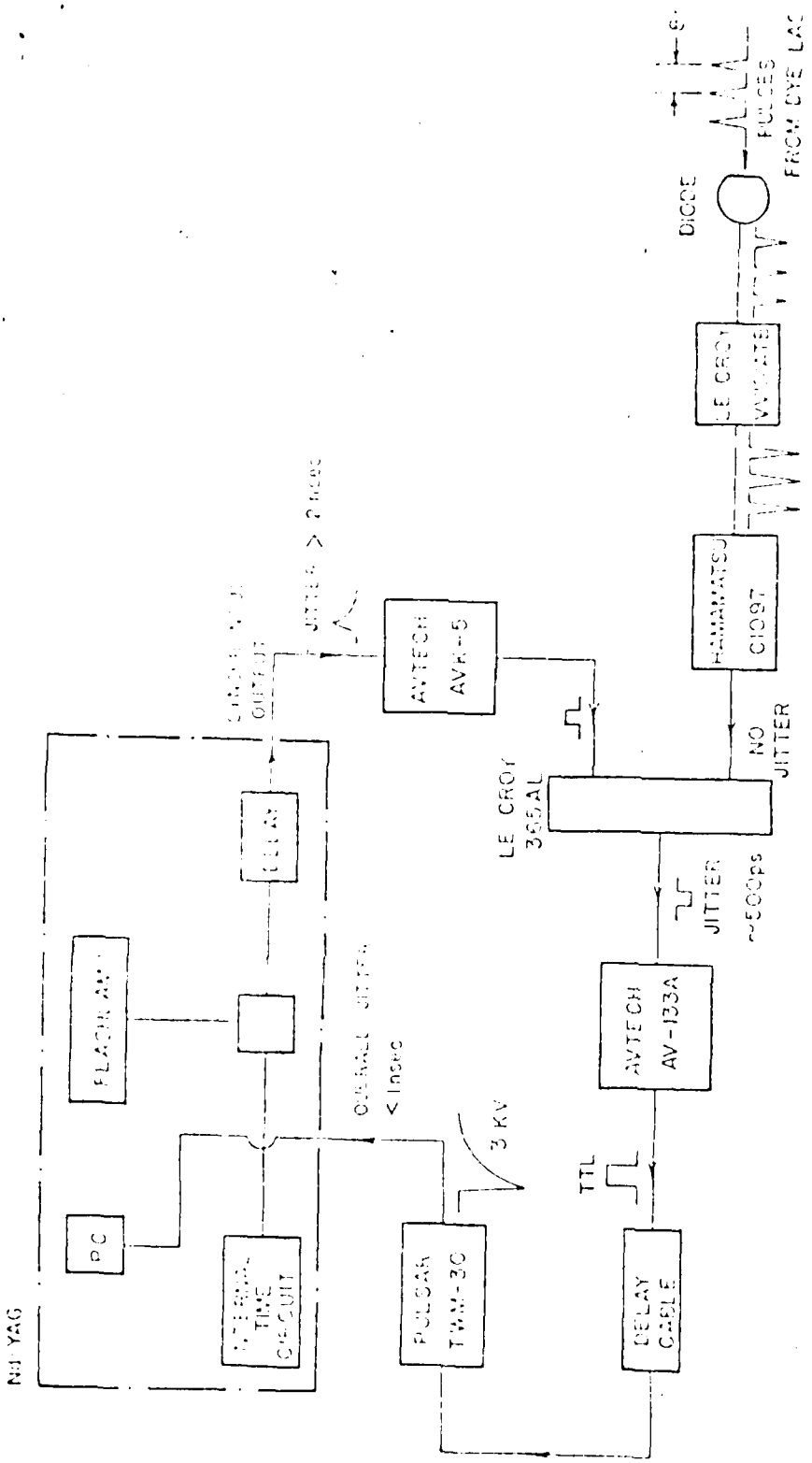


Fig. 2 Schematic diagram of the electronics used to trigger the amplifier

cell. There are two advantages in this design. First the system is immune to the jitter of the Nd:Yag electronics, since the pulse only opens the logic unit. The triggering occurs on the dye laser pulse which has practically no jitter. Second the electronics make use as much as possible of the internal electronics of the Nd:Yag laser with considerable savings in the overall cost.

The Hamamatsu delay unit is used to bring the two electronic pulses (from the Nd:Yag and the dye laser) to overlap at the coincidence unit. The overlap of the optical pulses at the first amplification stage is accomplished by adjusting the delay cable. The overlapping at the other two stages is insured because the path-length of the two beams are equal, see fig. 1.

B. Luminescence Measurements by Population Mixing Technique

We have continued the studies of the recombination of photo-generated carriers in semiconductors by population mixing. For a detailed description of this technique that was developed in our laboratory see interim progress report dated 1981. In this technique the pulse train is divided by a beam splitter into two pulse trains which are modulated at frequencies ω_1 and ω_2, respectively. A variable delay is added to one of the beams. Both beams are focused by a lens onto the sample. The fluorescence is collected and focused on the slit of a monochromator and detected by a photo-multiplier and a lock-in amplifier, set at the difference frequency $\omega_1 - \omega_2$. In this way only the fluorescence signal produced by the mixing of the populations generated by the two pulse trains

is detected. The two pulses create free carrier populations n(t) and n(t+τ), respectively. The detected signal is

$$I_s(\tau) \propto \int_{-\infty}^{\infty} n(t+\tau)\, n(t)\, dt$$

If a single exponential decay time T of the carrier is assumed, we obtain

$$I_s(\tau) \propto \exp\left(-\frac{|\tau|}{T}\right)$$

The correlation function provides information about the relaxation of the photogenerated carriers. The method has subpicosecond time resolution limited only by the duration of the pulse.

The technique has been applied to samples of p-GaAs and CdSe. We have measured the recombination kinetics in CdSe at the exciton line. The kinetics of recombination can be fitted to a double exponential of 35 and 50 psec. Research on a temperature dependence of relaxation times is in progress.

C. Luminescence Measurements from Ferromagnetic and Layered Semiconductors

Our work on steady state fluorescence of $CdCr_2Se_4$ has determined the location of the fundamental energy gap at 1.8 ev. This has been a matter of controversy for fifteen years.

The photoluminescence relaxation decay time of the band to band transition in $CdCr_2Se_4$ was measured from 4 K to 250 K. The

decay time was measured to be ~45 ps at 4 K which monotonically decreased to < 2 ps at 250 K. The integrated intensity of the steady state photoluminescence from $CdCr_2Se_4$ was measured from 15 K to 220 K. The intensity increased as the temperature decreased and increased quadratically with the excitation intensity. The temperature dependence of the relaxation decay time is theoretically fitted to the temperature dependence of the integrated intensity.

Photoluminescence spectra from $CdCr_2Se_4$ has been measured at low temperatures. When the temperature was lowered below 70 K, an extra peak was observed on the higher energy side of the band to band luminescence transition. The energy difference between the two peaks was measured to be 3.5 mev. The emission was attributed to the transitions from the split S-conduction bands.

Changes in photoluminescence spectra of the magnetic semiconductor $CdCr_2Se_4$ at 80 K were measured at high picosecond laser pulse intensities from 0.3 to 20 GW cm^{-2}. Evidence is presented for the formation of the electron hole plasma. Theoretical fitting of the spectral data determined that $CdCr_2Se_4$ is a direct band gap semiconductor.

D. Relaxation of Photogenerated Hot Carriers in Semiconductors by Time-resolved Spectroscopy

Free carrier relaxation times in GaAs have been shown to increase as the free carrier density increases by intense picosecond photogeneration. We have measured the hot photoluminescence kinetics of GaAs under intense picosecond excitations (10^{28} photons/cm^2 sec). A slow rise time of the near bandedge luminescence has been

observed arising from a slowed cooling of the electron distribution. The slowed kinetics can be attributed to the high optical phonon density and screening of the electron-phonon interaction. There are two theories which can explain the relaxation time of photogenerated hot electrons when the concentration of the hot carriers is very high in the sample GaAs. One is the screening of electron-lattice interaction leading to the emissions of LO-phonons from electrons. The other is the heated LO-phonon effect on the slowed relaxation time of the hot electrons. We have shown by comparing our experimental results to the two theories, that screening of the electron-phonon interaction is responsible for the observed slowing.

We have also measured time-resolved absorption and emission spectra of GaSe. The absorption measurements indicate that the dominant relaxation process for hot carriers in GaSe is the emission of non-polar optical phonons $A'^{(1)}_1$ from holes. In addition the deformation potential between holes and non-optical phonons is reduced five times when the carrier density reaches 10^{19} carriers/cm^3. The emission measurements support this by showing that the risetime of the luminescence increases when the carrier density increases. This indicates that screening effect of emission of optical phonons on the relaxation of hot carriers occurs at high carrier density.

The photoluminescence spectra of gallium selenide consist of two bands at both 77 K and 300 K. The lower energy part of the spectra is associated with stimulated emission. The higher energy part of the spectra is assigned to the exciton-electron (hole) scattering process at lower excitation intensity and to the electron hole plasma at higher excitation intensity. From the analysis of the high energy

tails of the higher energy part of the emission spectra, the Mott transition from the exciton state to the plasma state at 300 K was obtained when the excitation intensity is about (1.70 $\pm$ 0.25) 10^8 Wcm^{-2}.

The time resolved photoluminescence kinetics of GaSe has been measured at room temperature to determine screening of optical phonon-hole interaction. The risetime of the spontaneous emission band is within the response time of the picosecond laser pulse duration and streak camera ($\lesssim$ 20 ps) when the photogenerated carrier density is below $5x10^{18}$ cm^{-3} and increases to a measurable value when the carrier density is above $5x10^{18}$ cm^{-3}. The increase in the risetime of the spontaneous emission band arises from the screening of nonpolar optical phonon emission from hot photogenerated carriers due to high photogenerated carrier density. These measurements support our absorption kinetics.

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